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TOPICAL MEETING ON

MULTIPLE EXCITATIONS OF ATOMS

TECHNICAL DIGEST

OCTOBER 20-22, 1986
SEATTLE, WASHINGTON

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The, ILS-II, was held in Seattle from October 20-24 concurrently with the 1986 OSA ANnual Meeting, the Topical Meeting on Multiple Excitation of Atoms, and the Workshop on Optical Fabrication and Testing-all grouped together at the Seattle Center under the banner of the Coordinated Conference on Optical and Laser Science and Technology.

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TOPICAL MEETING ON

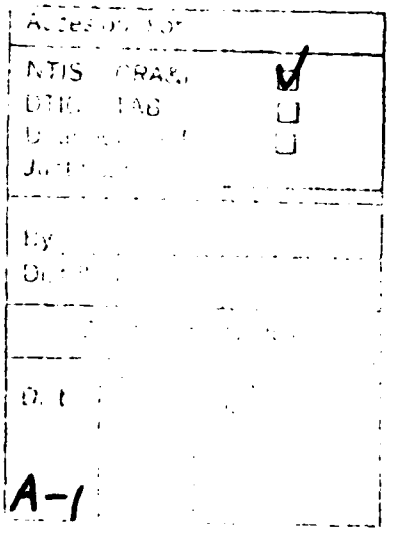
MULTIPLE EXCITATIONS OF ATOMS

Summaries of papers presented at the
Multiple Excitations of Atoms Topical Meeting

October 20–22, 1986

Seattle, Washington

Conference Edition



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SUNDAY, OCTOBER 19, 1986

ALKI ROOM

12:00 M–7:30 PM REGISTRATION

5:30 PM–7:30 PM REFRESHMENTS

MONDAY, OCTOBER 20, 1986

MERCER V

7:45 AM–8:20 AM CONTINENTAL BREAKFAST

MERCER III

8:20 AM OPENING REMARKS
William E. Cooke, *U. of Southern California, Meeting Cochair*

II-MA ABOVE THRESHOLD EXCITATION AND IONIZATION: 1
William E. Cooke, *U. of Southern California, Presider*

8:30 AM
II-MA1 Angular Distribution of Photoelectrons from Above-Threshold-Ionization (ATI) of Xenon Atoms, D. Feldmann, *U. Bielefeld, F. R. Germany.* The range of angular momenta of continuum states involved in ATI has been determined at the four harmonic wavelengths of a ND-YAG laser. (p. 2)

9:00 AM
II-MA2 Study of Ponderomotive Forces and Above-Threshold Multiphoton Ionization by Angle Resolved Photoelectron Spectroscopy, P. H. Bucksbaum, R. R. Freeman, T. J. McIlrath, M. Bashkansky, *AT&T Bell Laboratories.* Above threshold ionization and ponderomotive forces are studied using photoelectron energy, intensity, and angular distributions from 1064-nm multiphoton ionization of rare gases. (p. 3)

9:30 AM
II-MA3 Excess-Photon Ionization and Dressing of Continuum States, H. B. van Linden van den Heuvell, H. G. Muller, *FOM-Institute for Atomic & Molecular Physics, The Netherlands.* Excess-photon ionization is considered, both in a situation where the ionization and excess absorption are due to one field, and where ionization and continuum-continuum transitions are due to different fields. (p. 5)

9:50 AM COFFEE BREAK

MONDAY, OCTOBER 20, 1986 — Continued

10:15 AM
II-MA4 Resonantly Enhanced Multiphoton Ionization Photoelectron Angular Distributions for Xenon, P. R. Blazewicz, *Yale U.;* R. N. Compton, J. A. D. Stockdale, John C. Miller, *Oak Ridge National Laboratory.* Resonantly enhanced multiphoton ionization and above-threshold ionization photoelectron angular distributions via the $6s(3/2)_1^o$ and $6s'(1/2)_1^o$ resonant intermediate states have been studied. (p. 7)

10:35 AM
II-MA5 Monte Carlo Description of Photoionization, George A. Kyrala, *Los Alamos National Laboratory.* Photoionization of a hydrogenic atom in an intense electromagnetic field utilizing a classical mechanical description is introduced. The effects of polarization of the electromagnetic field are presented. (p. 9)

10:55 AM
II-MA6 The Ionization Threshold Problem in Intense Field Multiphoton Ionization of Atoms, Liwen Pan, Bala Sundaram, Lloyd Armstrong, Jr., *Johns Hopkins U.* We study the properties of final state wavefunctions in intense field multiphoton ionization of atoms, and the role of the ponderomotive potential. (p. 11)

11:15 AM
II-MA7 Multiphoton and Above-Threshold Ionization, Shih-I Chu, *U. Kansas.* Nonperturbative Floquet methods for intensity-dependent threshold shift, multiphoton and above-threshold ionization of atomic hydrogen in intense monochromatic/polychromatic laser fields are presented. (p. 13)

11:35 AM LUNCH BREAK

II-MB ABOVE THRESHOLD EXCITATION AND IONIZATION: 2
Steve J. Smith, *JILA, Presider*

1:30 PM
II-MB1 Theoretical Aspects of Above-Threshold Absorption, Robin Shakeshaft, *U. Southern California.* Some theoretical approaches to the description of above-threshold absorption are reviewed. (p. 16)

2:00 PM
II-MB2 Quantum Optical Approaches to the Problem of Atoms and Electrons in Intense Fields, J. H. Eberly, *U. Rochester.* We show how an application of quantum optic principles leads to some tentative conclusions regarding the behavior of atoms and electrons in intense radiation fields. (p. 17)

2:30 PM
II-MB3 Peak Suppression in Above-Threshold Ionization. P. Agostini, G. Petite, F. Yergeau, *CEN Saclay, France.* After a brief review of the experimental and theoretical material, we present and discuss new experimental results obtained in a regime free of space charge effects, using a magnetic collection and time-of-flight technique. (p. 18)

2:50 PM
II-MB4 Atomic Behavior in Intense High-Frequency Laser Fields, M. Gavrila, M. J. Offerhaus, M. Pont, *FOM-Institute for Atomic & Molecular Physics, The Netherlands;* J. Z. Kaminski, *Warsaw U., Poland;* J. Van de Ree, *Eindhoven U. Technology, The Netherlands.* A recently developed theory was applied to calculate nonperturbative levels, scattering and multiphoton transitions of the electron-nucleus system in intense, high-frequency laser fields. (p. 19)

3:10 PM COFFEE BREAK

3:30 PM
II-MC POSTDEADLINE PAPERS

SHAW ROOM

8:00 PM–11:00 PM
OSA SPECTROSCOPY TECHNICAL GROUP MEETING

A panel discussion will be held concerning the question, "Does the physics of atoms in strong AC fields bear any resemblance to the atomic physics that we all know and love?" Thomas J. McIlrath, *Moderator.*

MERCER V

7:45 AM–8:30 AM CONTINENTAL BREAKFAST

MERCER III

II-TuA TWO ELECTRON SYSTEMS
Robert N. Compton, *Oak Ridge National Laboratory, Presider*

8:30 AM
II-TuA1 Multiple Excitation of Atomic Electrons, Chris H. Greene, *Louisiana State U.* Extensive development of nonperturbative theoretical methods for handling electron correlations is needed to deal with doubly and triply excited states of atoms. Recent progress is described. (p. 22)

9:00 AM
II-TuA2 Doubly Excited Atoms, T. F. Gallagher, *U. Virginia.* A survey of doubly excited atoms includes such topics as their unique properties and different ways in which they may be produced or studied. (p. 24)

9:30 AM
II-TuA3 Pair Description of Two-Electron States, A. R. P. Rau, *Louisiana State U.* A consistent description of doubly excited states in terms of a pair of electrons rather than individual particles is developed further. (p. 26)

9:50 AM COFFEE BREAK

Michele Crance, *Laboratoire Aime Cotton, France, Presider*

10:15 AM
II-TuA4 Wannier Two-Electron Ionization Ladder in Many-Electron Systems: The $^1P^o$ Doubly Excited States in H^-, He, and Li^+, Yannis Komninos, Cleanthes A. Nicolaides, *National Hellenic Research Foundation, Greece.* We present a theory for the *a priori* identification and accurate calculation of the correlated wave functions of a special class of doubly excited states which lead smoothly to the Wannier state at threshold. (p. 30)

10:35 AM
II-TuA5 Regularities of Negative Ion Resonances, Charles W. Clark, Stephen J. Buckman, *Australian National U.* Some regularities of negative ion resonances are identified by isoionic comparisons. They shed light on Wannier ridge-and-valley aspects of multiply excited states. (p. 32)

10:55 AM
II-TuA6 Novel Motions in Highly Excited Two-Electron Atoms, Hubert Klar, *U. Kaiserslautern, F. R. Germany.* Novel attracting motions consisting of periodic, quasiperiodic and chaotic components predict a new resonance formation in doubly excited atoms. Also, double escape correlations are analyzed. (p. 34)

11:15 AM
II-TuA7 Inhibited Autoionization in Multicontinua Systems, V. Lange, G. Renz, U. Eichmann, A. Zetsche, W. Sandner, *U. Freiburg, F. R. Germany.* Experimental and theoretical studies of inhibited autoionization in multicontinua systems are presented. Applications include controlled population inversion of ions by excitation through autoionizing Rydberg states. (p. 35)

11:35 AM LUNCH BREAK

II-TuB MULTISTAGE IONIZATION
See Leang Chin, *Laval U., Presider*

1:30 PM
II-TuB1 Multiply Charged Ions and Energy Spectrum of Electrons in Multiphoton Ionization, L.-A. Lompre, G. Mainfray, *Centre d' Etudes Nucleaires de Saclay, France.* Formation of multiply charged ions and energy spectrum of electrons produced in the multiphoton ionization of rare gases obtained in a strong laser field are discussed. (p. 38)

2:00 PM
II-TuB2 Many-Electron Dynamics of Heavy Atoms in Intense Laser Fields: Effects of Screening and Correlation, Goran Wendin, Lars Jonsson, Anne L'Huillier, *Chalmers U. Technology, Sweden.* We discuss the role of collective effects in multiphoton-multielectron ionization of heavy atoms. We present results of an application to two-photon one-electron ionization of the 5*p*-shell of xenon in the 0.45–1.4 Ry photon energy region. Finally, we discuss briefly different aspects of many-electron effects in direct and stepwise multielectron radiation. (p. 40)

2:30 PM
II-TuB3 Time-Dependent Hartree Fock Theory of Multiphoton Ionization, Kenneth C. Kulander, *Lawrence Livermore National Laboratory.* The application of a time-dependent self-consistent field method to the interaction of a multielectron atom with intense laser fields is discussed. (p. 41)

FLAG PAVILION

3:00 PM–5:00 PM
II-TuC POSTER PREVIEWS/REFRESHMENTS

Poster papers and overhead transparencies of selected oral presentations will be available for preview.

TUESDAY, OCTOBER 21, 1986 — Continued

6:00 PM–8:00 PM
II-TuC POSTER PRESENTATIONS

Poster papers and overhead transparencies of selected oral presentations. Authors will be in attendance.

II-TuC1 Dynamics in Intense Fields: Beyond the Dipole Approximation, Andre D. Bandrauk, O. P. Kalman, *U. Sherbrooke, Canada.* The dressed molecular Hamiltonian for molecules in strong electromagnetic fields is derived in the Bloch Nordsieck (BN) electric field (EF) representations beyond the dipole approximation. Examples of analytic and numerical field modified potentials are presented. (p. 44)

II-TuC2 Dressing to the Hydrogen Atom in an Extremely Intense Laser Field, M. Janjusevic, M. H. Mittleman, *City College of New York.* We have rederived an equation for the dressed bound state of the H atom. One immediate result is that the level shift is the ponderomotive potential plus smaller terms. The applicability of the theory to other atoms is discussed. (p. 47)

II-TuC3 Inverse Half-Bremsstrahlung in Multiphoton Ionization of Atoms in Intense Light Beams, Joseph Kupersztych, *CEN Saclay, France.* It is shown that absorption of the minimum number of photons needed to ionize an atom in a light beam of moderate intensity is no longer energetically possible when the external field is strong. However, subsequent absorption of a large number of photons by the emerging electron in the field of its parent ion can occur via multiphoton inverse half-bremsstrahlung and ionization is again possible. (p. 48)

II-TuC4 Final State Effects in Above-Threshold Ionization, W. Becker, M. O. Scully, *U. New Mexico;* R. R. Schlicher, *Max-Planck Institute for Quantum Optics, F. R. Germany.* We propose a simple model which exhibits most of the universal features encountered in the electron spectra of above-threshold ionization. (p. 49)

II-TuC5 Strong Field Laser Ionization of Model Atoms, M. S. Pindzola, *Auburn U.;* C. Bottcher, *Oak Ridge National Laboratory.* Finite element methods are used to solve the time-dependent Schrodinger equation for a 1-D square well potential subject to an arbitrarily strong electromagnetic field. (p. 51)

II-TuC6 General Relativistic Ponderomotive Force in a Moving Medium, Shitong Zhu, *Shanghai Institute of Optics & Fine Mechanics, China;* Wenda Shen, *Shanghai U. Science & Technology, China.* The general relativistic ponderomotive force is first derived. Previous results are modified. Unlimited ponderomotive force in a moving plasma with critical density is predicted. (p. 52)

II-TuC7 Classical Calculations of the Effects of High Brightness Laser Radiation on Photoelectron Angular Distributions, Don R. Kania, *Los Alamos National Laboratory.* A two-step model (photoionization and transport) has been developed to calculate the effects of high brightness laser radiation on photoelectron angular distributions. (p. 54)

II-TuC8 Model of Two-Electron Ionization based on Semiclassical Phase Space Averaging, Jan Mostowski, *U. Rochester;* Marek Trippenbach, *Warsaw U., Poland;* Cao Long Van, *Institute for Theoretical Physics, Poland.* Ionization of helium by a very strong laser is studied by phase space averaging of Newtonian trajectories, using the ground-state Wigner function at $t = 0$. (p. 56)

II-TuC9 Collective Dipole Excitation of Many-Electron Atoms by Intense Laser Pulse, Farhad H. M. Faisal, *U. Bielefeld, F. R. Germany.* Many-body random-phase analysis of atomic Hamiltonian is made; existence of laser driven collective excitation of atomic dipole operator is demonstrated; absorption cross-sections in noble gases are obtained. (p. 58)

II-TuC10 Electron Angular Distributions and Branching Ratios of the SrI $5p_{3/2}ns_{1/2}$ and $5p_{1/2}ns_{1/2}$ $J = 1$ Autoionizing States, Y. Zhu, E. Y. Xu, T. F. Gallagher, *U. Virginia.* The energy and angular distributions of electrons ejected from the SrI $5p_{3/2}ns_{1/2}$ and $5p_{1/2}ns_{1/2}$ $J = 1$ autoionizing states have been measured and a six-channel MQDT analysis is presented. (p. 60)

II-TuC11 Photoelectron Spectrum Resulting from Autoionizing State Resonance, Lee-Geng Zhao, Zhi-Zhan Xu, *Shanghai Institute of Optics & Fine Mechanics, China.* The photoelectron spectrum produced by autoionizing state resonance in the continuum has been studied theoretically by employing the resolvent operator method. (p. 62)

II-TuC12 Multiphoton Isolated Core Excitation, J. G. Story, L. D. Van Woerkom, W. E. Cooke, *U. Southern California.* A closed form expression for multiphoton core excitation is presented. Guidelines are presented for maximizing an n photon core excitation. (p. 64)

II-TuC13 Observation and Analysis of the Autoionizing Spectra of Sr, Sufen Hu, Sen Zhang, Jie Lu, Jizhen Qiu, Jiazhen Sun, *Zhejiang U., China.* The Sr($5p_{1/2}ns$), and $(5p_{1/2}nd)_{1,3}$ autoionizing levels have been measured using the polarized laser multiple excitation approach. Some results have been analyzed. (p. 66)

TUESDAY, OCTOBER 21, 1986 — Continued

II-TuC14 Multiphoton Ionization of Beryllium, J. Wen, J. C. Travis, T. B. Lucatorto, C. W. Clark, *U.S. National Bureau of Standards.* We have measured isotope shifts and hyperfine structure of singly and doubly excited states of beryllium and have studied line shapes associated with resonant multiphoton ionization. (p. 68)

II-TuC15 Electric Field Effects on Doubly Excited Autoionizing States, D. E. Kelleher, E. B. Salomon, J. W. Cooper, C. W. Clark, *U.S. National Bureau of Standards.* We summarize electric-field effects observed on autoionizing resonances: field-induced autoionization and interferences, and ion-core anisotropy effects on Rydberg-Stark manifolds. (p. 70)

II-TuC16 Double Electron Excitation of Lithium by Electron Impact, R. Srivastava, *U. Roorkee, India;* W. Williamson, Jr., *U. Toledo.* The electron impact total cross section for the double electron excitation of lithium using the distorted wave theory has been calculated. The results are reported for the Li($^2S \rightarrow {}^4P$) transition. The distorted wave model used includes distortions due to the static, polarization, and exchange effects appropriately in both the incoming and outgoing distorted waves. (p. 72)

6:00 PM–8:00 PM RECEPTION

WEDNESDAY, OCTOBER 22, 1986

MERCER V

8:30 AM–9:00 AM CONTINENTAL BREAKFAST

MERCER III

II/III-WA JOINT SYMPOSIUM OF THE TOPICAL MEETING ON MULTIPLE EXCITATIONS OF ATOMS AND THE INTERNATIONAL LASER SCIENCE CONFERENCE
Thomas J. McIlrath, *U. Maryland, Presider*

9:00 AM
II/III-WA1 Multiphoton Excitation and Ionization of Atoms, H. Jara, U. Johann, T. S. Luk, I. A. McIntyre, A. McPherson, A. P. Schwarzenbach, K. Boyer, C. K. Rhodes, *U. Illinois at Chicago.* The results of the subpicosecond studies at 248 nm are discussed. (p. 76)

9:30 AM
II/III-WA2 Laser Spectroscopy of Core-Excited Levels of Neutral Rubidium, S. E. Harris, J. K. Spong, J. D. Kmetec, K. D. Pedrotti, S. C. Wallace, J. F. Young, *Stanford U.* We describe first experimental results of a new technique for obtaining level positions, autoionizing linewidths, and oscillator strengths of core-excited levels and transitions. (p. 79)

10:00 AM
II/III-WA3 Overview of Above Threshold Ionization: There is Only One Physics, R. R. Freeman, *AT&T Bell Laboratories.* We review the recent work on ATI and show that the phenomena can be satisfactorily understood in the context of contributions from related fields. (p. 81)

10:30 AM
II/III-WA4 Multiple Excitation and Ionization of Atoms by Strong Lasers: Is There Any New Physics?, P. Lambropoulos, *U. Southern California.* A theoretical analysis of sequential (stripping) and direct ionization processes is employed in the description of multiple electron ejection. A formalism and calculations for the description of double electron ejection including laser intensity effects are also presented. (p. 82)

11:00 AM CLOSING REMARKS
Thomas J. McIlrath, *U. Maryland, Meeting Cochair*

PACIFIC SCIENCE CENTER

6:00 PM–8:00 PM CONFERENCE RECEPTION

MONDAY, OCTOBER 20, 1986

MERCER III
8:30 AM–11:35 AM

II-MA1–7

ABOVE THRESHOLD EXCITATION AND IONIZATION: 1

William E. Cooke, University of Southern California, *Meeting Cochair*

Angular Distribution of Photoelectrons from Above-Threshold-Ionization (ATI) of Xenon Atoms

D. Feldmann
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Federal Republic of Germany

The range of angular momenta of continuum states involved in ATI has been determined at the four harmonic wavelengths of a Nd-YAG laser.

Study of ponderomotive forces and above-threshold multiphoton ionization by angle resolved photoelectron spectroscopy

P. H. Bucksbaum, R. R. Freeman, T. J. McIlrath[a], M. Bashkansky[b]
AT&T Bell Laboratories
Murray Hill, NJ 07974

In investigations of above-threshold ionization (ATI)[1], we have shown that ponderomotive forces dominate photoelectron angular distributions at high laser intensities. The experiments employ 1064 nm 100 psec laser pulses or their harmonics, to ionize xenon and other rare gases, with peak intensities up to 10^{14} w/cm^2.

As a photoelectron leaves the tight focus of a laser beam where it was produced, it scatters due to ponderomotive forces proportional to the gradient of the laser intensity.[2] At low intensities, the electrons are emitted along the polarization axis,[3] but as the laser peak intensity is raised, the lower energy electrons become more isotropic, reflecting ponderomotive scattering from the focus (figure 1). In addition, the spatial mode can dramatically affect the distributions. Trajectory calculations agree well with these observations.

Figures 2a,b,c show the electron spectrum for linearly polarized light, observed along the polarization axis for different intensities. The threshold ionization processes (0.58 and 0.69 eV) are suppressed due to the shift of the atomic ionization energy in intense light fields.[4] Ionization with circularly polarized radiation can dramatically alter the ATI spectra by suppressing low energy electrons (figure 2d,e). A simple model, based on angular momentum absorption and suppression of the overlap with low energy final states by centrifugal repulsion, agrees well with these observations.[5]

[a] On leave from IPST, University of Maryland
[b] Physics Department, Columbia University

1. P. Kruit, J. Kimman, H.G. Muller and M.J. van der Wiel, Phys. Rev. A *28*, 248 (1983).
2. a: T.W.B. Kibble, Phys. Rev. *150*, 1060 (1966); b: J.H. Eberly, Progress in Optics, *7*, 359 (1969); c: P. Avan, C. Cohen-Tannoudji, J. Dupont-Roc and C. Fabre, J. de Physique *37*, 993 (1976); d: M. H. Mittleman, *Theory of Laser-Atom Interactions*, (Plenum, NY, 1982).
3. H. J. Humpert, H. Sckwier, R. Hippler and H. O. Lutz, Phys. Rev. A *32*, 3787 (1985).

4. H. G. Muller, A. Tip and M. J. van der Wiel, J. Phys. B *16*, L679, (1983); H. G. Muller and A. Tip, Phys Rev A *30*, 3039 (1984); M.H. Mittleman, J. Phys. B *17*, L351 (1984); A. Szoke, J. Phys. B, *18*, L427 (1985). This was observed in Ref. 1, and in L. A. Lompre, A.L'Huillier, G. Mainfray and C. Manus, J. Opt. Soc. Am. B *2*, 1906 (1985).

5. P. H. Bucksbaum, M. Bashkansky, R. R. Freeman, and T. J. McIlrath, Phys. Rev. Letters *56*, June 19 issue, in press (1986).

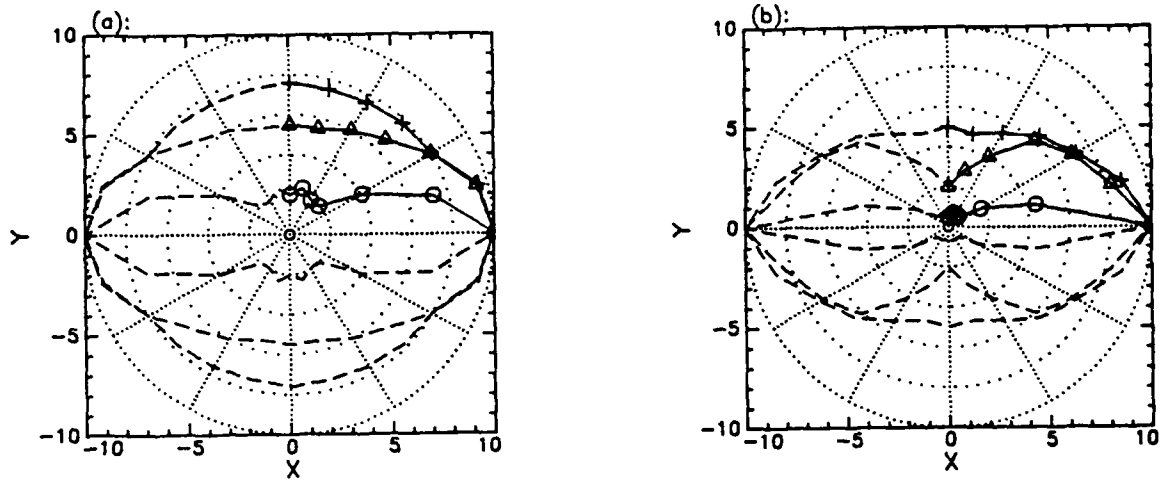

Figure 1: Photoelectron angular distributions in the polarization plane, for linear polarization along 0°; o, Δ, + correspond to 0.7, 1.4, 11.2 x10^{13}w/cm^2, respectively. a: 1.8 eV S=1 electrons; b: 3.0 eV S=2 electrons.

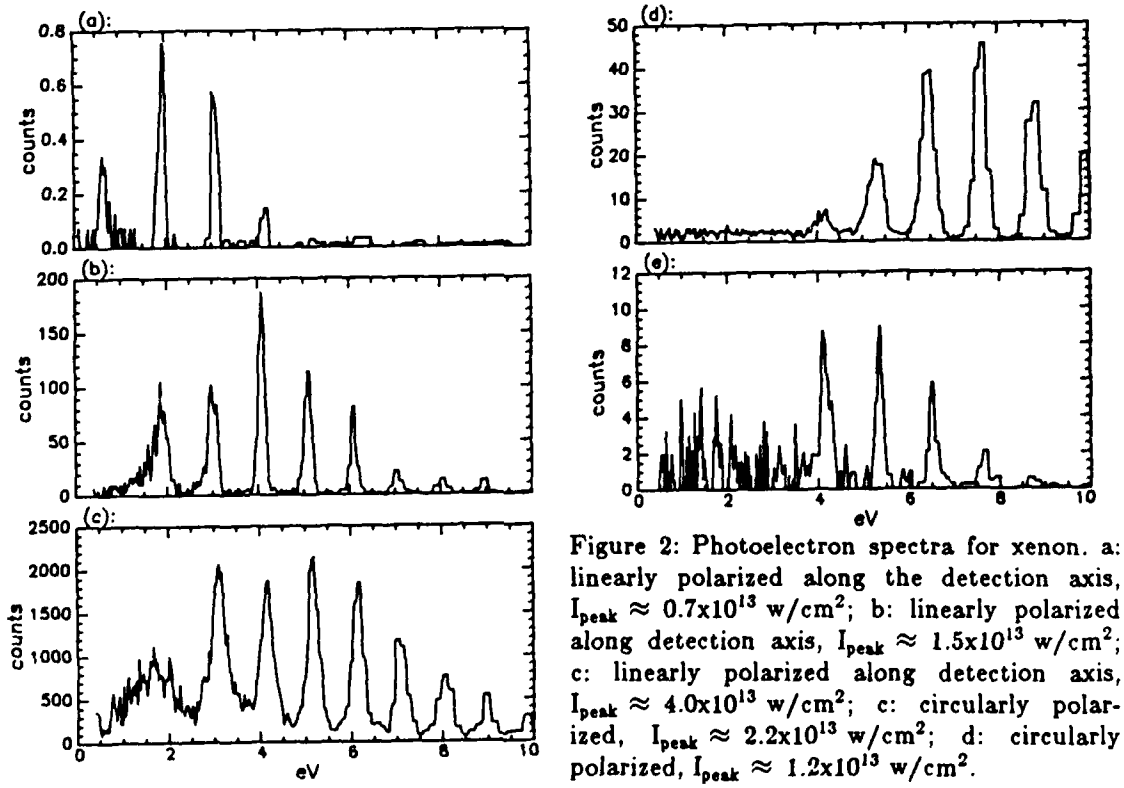

Figure 2: Photoelectron spectra for xenon. a: linearly polarized along the detection axis, $I_{peak} \approx 0.7 \times 10^{13}$ w/cm^2; b: linearly polarized along detection axis, $I_{peak} \approx 1.5 \times 10^{13}$ w/cm^2; c: linearly polarized along detection axis, $I_{peak} \approx 4.0 \times 10^{13}$ w/cm^2; c: circularly polarized, $I_{peak} \approx 2.2 \times 10^{13}$ w/cm^2; d: circularly polarized, $I_{peak} \approx 1.2 \times 10^{13}$ w/cm^2.

Excess-Photon Ionization and Dressing of Continuum States.

H.B. van Linden van den Heuvell and H.G. Muller
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Among the features of (quasi-)one-electron atoms in a strong radiation field, the process of excess-photon ionization is one of the most spectacular. For a short time an electron is coupled both to the remaining core of an atom and to a radiation field. This leads to a gain af kinetic energy of the electron. This gain is not possible if only one of the two couplings is present.

A recent suggestion[1] is followed here to use the name excess-photon ionization (EPI) for this process, rather than the more traditional but unfortunately somewhat confusing name of above threshold ionization (ATI).

In this contribution we would like to compare two classes of experiments: EPI-experiments on the one hand (e.g. the experiment reported by Kruit et al.[2]) and normal multiphoton ionization (where the ionization takes place with the smallest possible number of photons in a relatively weak field) plus a strong radiation field deforming the continuum states, on the other hand. This situation can of course only be arranged if the strong dressing field (ω_2) has a substantially lower photon energy than the ionizing radiation field (ω_1).

From a theoretical point of view the experiment with two radiation fields is easier to describe than EPI since in that case the total process can be separated in an ionization step and a dressing step. In quite a lof of descriptions for EPI this separation is also assumed. However, in the case of two radiation fields, the separation between ionization and dressing does not have to be assumed. In fact, this problem is excatly solvable if the energy of the ionized electrons is much larger than the photon energy of the radiation field. This would be the case for the series $4\omega_1 \pm N\omega_2$ in figure 1. however this series is complicated by the fact that also another path leads to the same series, namely the one which involves an ω_2 photon emission while the electron is still bound. This description in terms of side bands has a large analogy with the one of Kroll and Watson[3] for free-free scattering.

References:

1) B. Shore (1985) and also suggested at the Workshop 'Atoms in strong radiation fields', Orsay (1986).
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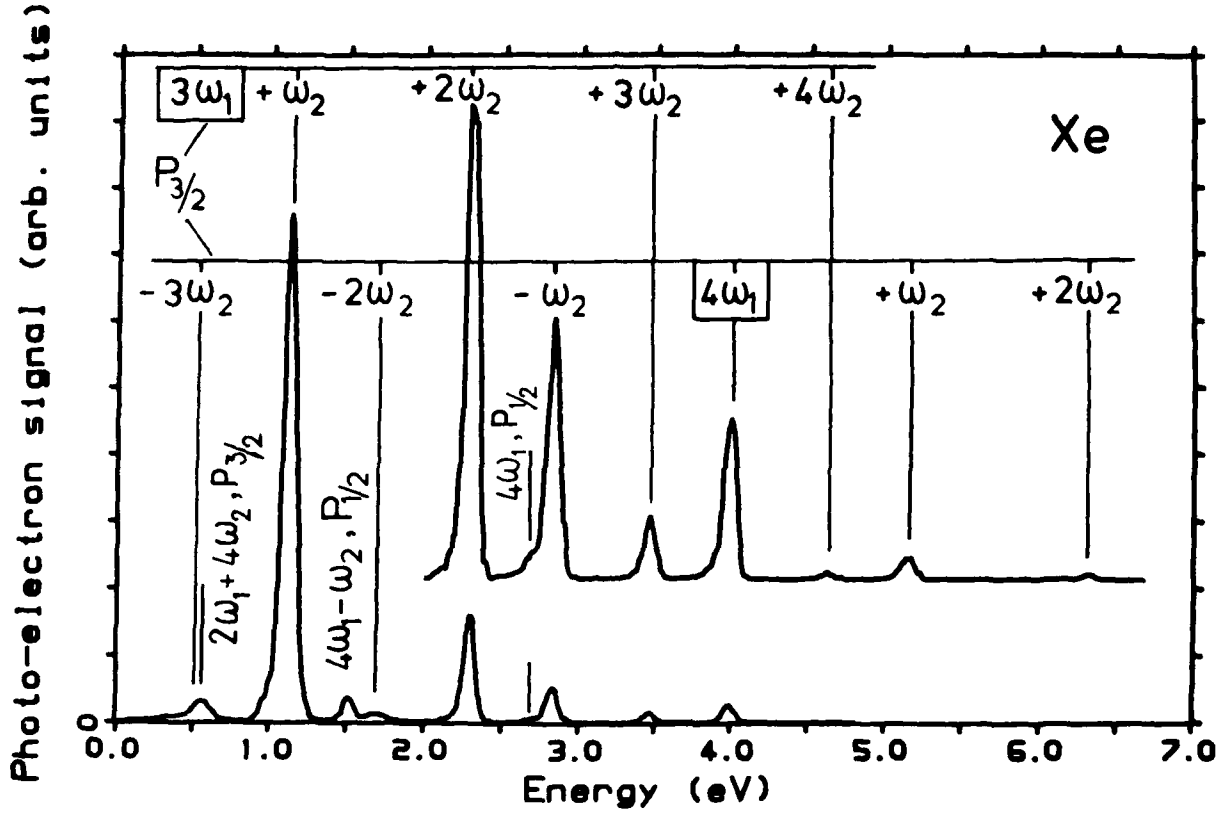

figure 1.: Spectrum of photoelectrons after photoionization of xenon with 300 nm light of intensity $\approx 10^{10}$ *W/cm*^{2} *in an additional laserfield of 1064 nm and* $7 \cdot 10^{11}$ *W/cm*^{2}.

Resonantly-Enhanced Multiphoton Ionization Photoelectron Angular Distributions for Xenon

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Multiphoton ionization (MPI) of Xe atoms resonantly enhanced through the three-photon allowed $6s[3/2]_1^o$ and $6s'[1/2]_1^o$ states has been extensively studied for high- and low-gas densities (see, e.g., Ref. 1). Limited data exist on the angular distributions of the photoelectrons ejected under these conditions.[2,3]

In this work we report energy-resolved angular distributions from ionization through pathways resonant at the three-photon level with either the $6s[3/2]_1^o$ or $6s'[1/2]_1^o$ level in Xe.

Briefly, the output of a pulsed, amplified dye laser (Quanta Ray PDL-2) pumped by a Nd:YAG laser (Quanta Ray DCR-2) was polarization purified with a Glan-air prism and focused with a 38-mm or 50-mm focal length lens to give a power density ~10^{10} W/cm^2. Xenon gas was introduced into the vacuum chamber with a pulsed nozzle (Laser Technics). The plane of polarization of the dye laser could be rotated continuously by rotating a double Fresnel rhomb with a stepping motor drive. The photoelectrons are energy analyzed with a 7.3-cm mean radius spherical-sector analyzer operated in fixed pass-energy mode.

Five photons are required to ionize Xe through the 6s state (3 + 2 ionization) leaving the ion in either the $^2P_{3/2}$ or $^2P_{1/2}$ core and producing two corresponding photoelectron peaks. The high-energy electrons have an angular distribution consisting of maxima at 0 and π radians with smaller submaxima at $\pi/2$ and $3\pi/2$. These electrons are due to ionization which preserves the $^2P_{3/2}$ core of the 6s state. The low-energy electrons have an

angular distribution which is less steep with maxima at 0 and π only and result from a "core-changing" ionization. These distributions might be expected to be influenced by states near the four-photon level. An a.c. Stark broadening of the 6s ionization is easily observed,[3] but we see no change in the angular distributions as we tune over the profile which is broadened to 5-7 Å under our conditions.

(3 + 1) ionization through the 6s' state exceeds the energy of the Xe^+ $^2P_{3/2}$ state only and produces one major photoelectron peak. This is also a "core-changing" ionization. The angular distribution of these photoelectrons is markedly isotropic. It is very similar to an angular distribution calculated for two-photon excitation of a J=2 autoionizing resonance in Xe.[4] The 6s' photoelectron spectrum also includes two weaker peaks due to absorption of a fifth photon leaving the ion in either core. Much weaker above threshold ionization peaks have also been seen in the 6s photoelectron energy spectrum.

Research is sponsored by the Office of Health and Environmental Research, U.S. Department of Energy under contract DE-AC05-840R21400 with Martin Marietta Energy Systems, Inc.

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Monte Carlo Description of Photoionization.

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The interaction of an atom with a very intense external electromagnetic field is one of considerable current interest, particularly when the magnitude of the external applied fields are of the order of, or stronger than the internal atomic field experienced by the atomic electrons.In that case perturbation treatments of the interaction should be very difficult, if not incorrect. One reaches the point where one has to decide between which picture , among many, would be used for the interaction. Instead of using a multiphoton picture for the interaction, I chose to investigate a description of the interaction utilizing classical mechanics.

In this description one treats the electromagnetic field as an external force on the electron, one neglects the electron radiation field and its reaction on the external filed, one treats the hydrogen atom as a Kepler problem, and then one follows the orbits of the electron in the combined fields of the electromagnetic wave and the coulomb potential.

However, in order to reproduce the behavior of the hydrogenic atom, the initial conditions of the hydrogenic electron are chosen from a microcanonical distribution defined by the quantum mechanical probability distributions for the selected quantities. The procedure is quite similar to the successful Monte-Carlo method for classical collisions between electrons and atoms(ref 1).

Thresholds in the photoionization probability as a function of the external field intensity have been found. The threshold intensity depends on the electrons initial angular momnetum and eccentricity. Results will be presented that will show some of the classical orbits as well as the angular distribution of the photoelectron that are ejected.

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The Ionization Threshold in Intense Field Multiphoton Ionization of Atoms

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In recent studies of intense field above-threshold multiphoton ionization of atoms, the ponderomotive potential is introduced to explain the suppression of lower energy photoelectrons.[1] This explanation, which we will refer to as PPT (Ponderomotive Potential Theory), predicts an increase of ionization potential with increasing laser intensity. When using linearly polarized laser light with an intensity of about $10^{13}W/cm^2$, and a wavelength of 1064 nm, this increase of the ionization potential will be about an eV.

However, many experimental results on the total ion-yield as a function of intensity show very good agreement with the power law dependence predicted by lowest order perturbation theory. This agreement is good even when the laser intensity reaches $10^{15}W/cm^2$.[2] At this intensity, however, the ionization potential would be shifted by 100 eV according to the prediction of the PPT, which seemingly should cause a change in the power law dependence of the ionization.

We show that the crucial point in resolving these apparently contradictory results lies in choosing final states which are most appropriate to describe the experimental measurements. Volkov-type states are appropriate for studies of the photoelectron spectrum.[3] Simpler formulations in terms

of field-free states suffice for calculating ionization probabilities.

We have investigated the process of multiphoton ionization in an intense field by solving a quantized atom-field model in which the basis of final states includes all states necessary to allow an asymptotic Volkov-like behavior. By regrouping final states, we are able to provide physical interpretations of the processes involved. We discuss the role of the ponderomotive potential.

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Multiphoton and Above-Threshold Ionization

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Recently a number of advances have been made regarding the developments of semiclassical Floquet theories for ab initio nonperturbative treatments of intense field multiphoton processes.[1,2] These include multiphoton excitation (MPE) and dissociation (MPD) of molecules,[1] multiphoton ionization (MPI) of atoms,[1] many-mode Floquet theory,[1,2] SU(N) dynamic symmetries and symmetry breaking,[2] charge exchange in laser fields[2] and nonlinear optical processes etc.[3]

In this paper, we shall present our recent work in the area of intense field multiphoton and above-threshold ionization. In particular, we shall discuss: (i) L^2 non-Hermitian Floquet method for the study of threshold shift and above-threshold multiphoton ionization of atomic hydrogen in intense monochromatic laser fields;[4] (ii) two-color multiphoton ionization by means of non-Hermitian many-mode Floquet theory.[5] The most recent results and alternative nonperturbative techniques for the above-threshold ionization problem will also be discussed.[5]

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MONDAY, OCTOBER 20, 1986

MERCER III
1:30 PM–3:10 PM

II-MB1–4

ABOVE THRESHOLD EXCITATION AND IONIZATION: 2

Steve J. Smith, JILA, ***Presider***

Theoretical aspects of above-threshold absorption

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We will review computational and physical aspects of the ionization of an atom by N photons with N>P, where P is the minimum number of photons required to ionize the atom. In perturbation theory the matrix element, $M^{(N)}$, for N-photon absorption can be decomposed as $M^{+(N)} + M^{-(N)}$, which results from the decomposition of the stationary wave of the emergent photoelectron into its outgoing (+) and ingoing (-) parts. Each of the $M^{\pm(N)}$ are N-dimensional integrals over N radial coordinates r_i, i=1, ...,N. Although these integrals are not formally convergent for N>P, the integrand of $M^{+(N)}$ consists of functions that either decay exponentially or are outgoing waves for $r_i \sim \infty$. Hence the rotation $r_i \rightarrow r_i e^{i\theta}$, $0<\theta<\pi/2$, yields an integral representation of $M^{+(N)}$ with an exponentially decaying integrand, permitting direct and accurate numerical integration. A relation connecting $M^{-(N)}$ to $M^{+(N)}$ can then be used to compute $M^{(N)}$ for N>P with not much greater effort than for the case N=P.

Perturbation theory is expected to fail when N>P for moderately intense low frequency fields. Various models have been proposed to study multi-photon absorption in the non-perturbative region. A typical assumption underlying these models is that free-free transition matrix elements are insensitive to the energies of the continuum states. This assumption precludes the important physical effect that absorption is favored over emission. Several numerical approaches have been proposed, based on the Fourier expansion of the wavefunction in the time domain. We will review progress that has been made toward treating real systems.

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Title: Quantum Optical Approaches to the Problem of Atoms and Electrons in Intense Fields

Abstract: The principles of quantum optics suggest certain points of view in problems involving the interaction of intense radiation with atoms and electrons. These principles can frequently be used as a guide regarding the value of perturbation theory, the utility of dressed-state or Floquet methods, the presence or absence of significant electron-photon correlations, the relevance of Fermi's Golden Rule, and so on. They can also point to the existence of possible scaling relationships and assist in the qualitative interpretation of diverse data. We will attempt to show how an application of these principles leads to some tentative conclusions regarding the behavior of atoms and electrons in intense radiation fields.

Peak suppression in Above-Threshold-Ionization

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Recently, the Above-Threshold-Ionization (ATI) phenomenon has received much attention, in particular from theoreticians bringing up models to interpret the suppression of the low-energy part of the electron energy spectrum. In this respect, the reasons why some electron peaks should be suppressed can be classified in three groups: first, intensity effects which modify, in all models, the ATI probabilities and in some of them would induce strong Stark shifts of the ionization limit. Second, polarization effects: it has been proposed that in circular polarization, the high-angular-momentum final states, which are also the high-energy part of the spectrum, should dominate over the low-angular-momentum ones. Third, space charge effects, which could prevent the low-energy electrons from escaping from the interaction region. In this paper, after a brief review of the experimental and theoretical material, we present and discuss new experimental results obtained in a regime free of space charge effects, using a magnetic collection and time-of-flight technique. In xenon at 1064 nm, the main results are: (i) the first peak of the ATI spectrum is suppressed at 5×10^{12} W.cm^{-2} with linear polarization and at a lower value in circular polarization. For both polarizations, the peak suppression takes place at intensities much lower than the saturation intensity. For circular polarization, the effect of the centrifugal barrier which has been proposed recently is discussed, with the conclusion that it may enhance the peak supression which remain, fundamentally an intensity effect.

ATOMIC BEHAVIOR IN INTENSE, HIGH-FREQUENCY LASER FIELDS

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A theory was recently developed to describe electron-atom interaction in intense, high-frequency laser fields [1,2,3]. It applies to the intensities and frequencies already available from excimer lasers in the VUV [4], but extends beyond, to the X-ray range.

The radiation was assumed purely monochromatic (generalizations to a laser pulse are in progress). By a Floquet analysis, the Schrödinger equation was reduced to an infinite set of time-independent coupled equations for the components of the wave function. It was shown that in the high-intensity, high-frequency limit, these reduce to a single energy-eigenvalue type equation containing a "dressed" potential of axial symmetry (case of linear polarization). Consequently the atom is stable in this limit (although under circumstances strongly distorted) and, in the collision problem, only elastic scattering can occur. At high, but finite frequencies (next approximation), multiphoton transitions become possible and expressions were derived for their rates. The conditions required for the validity of these results were carefully investigated.

The theory was applied so far to one-electron systems. Exploratory calculations were carried out for laser-modified elastic scattering from Coulomb [5], and Yukawa potentials (submitted for publication). We now report on accurate results for the level structure and multiphoton ionization rates of atomic hydrogen, and on a coupled l-channel calculation for elastic scattering and free-free transitions in a Coulomb potential. Both cases reveal a picture different from that known at low and moderate frequencies. Thus, in Fig. 1 we show the α_o-dependence of the ground state of hydrogen ($\alpha_o = I^{1/2}\,\omega^{-2}$ a.u. is the only parameter entering this approximation). As apparent, a dramatic *decrease* of the ionization potential takes place at values of α_o now accessible experimentally. This contrasts to the situation in other radiation regimes and has far-reaching consequences on the ionization features (e.g. displacement of peaks due to ponderomotoric effects, etc.). The explicit ω- and I-dependence of the excess-photon ionization (EPI/ATI) peaks is obtained.

To illustrate the collision problem, we show in Fig. 2 the scattering angle dependence of the ratio $R = (d\sigma/d\Omega)/(d\sigma_c/d\Omega)$ of the laser-modified elastic cross section to the original Rutherford one. (The angles θ_i and ϕ characterize the orientation of the electron initial and final momenta with respect to the polarization vector and will not be defined here). At small angles "Coulomb

interference oscillations" appear, due to the departure of the dressed potential from the Coulomb form close to the nucleus. For larger α_0-values, the scattering can be strongly manipulated by varying the field.

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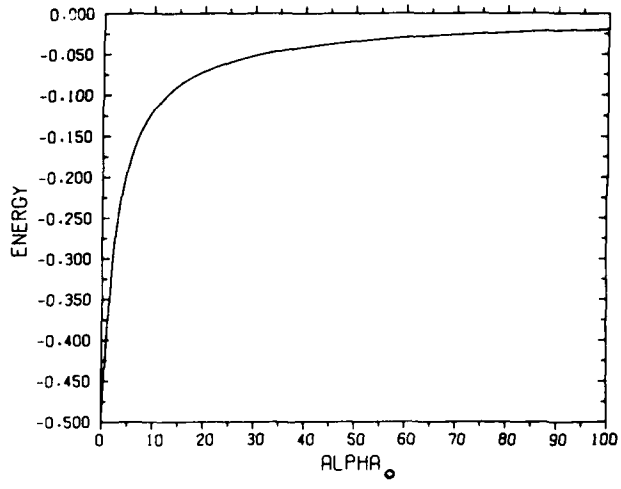

Fig. 1

α_0-dependence of ground state of hydrogen.

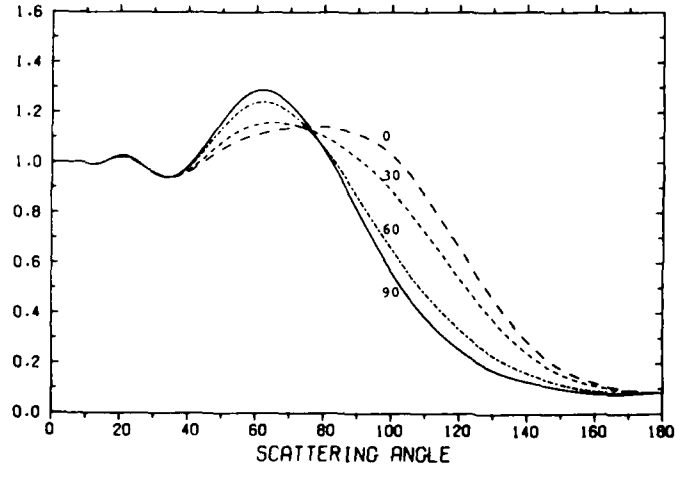

Fig. 2

Ratio R for the Coulomb potential. Energy 0.1 Ry, $\alpha_0 = 2$, $\theta_i = 90^o$, ϕ as indicated.

TUESDAY, OCTOBER 21, 1986

MERCER III
8:30 AM–9:50 AM

II-TuA1–3

TWO ELECTRON SYSTEMS

Robert N. Compton, Oak Ridge National Laboratory, *Presider*

Multiple Excitation of Atomic Electrons

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The strong interactions between simultaneously-excited electrons require nonperturbative theoretical techniques for their successful description. Two such nonperturbative techniques which have been pursued vigorously in recent years are the adiabatic treatment in hyperspherical coordinates and the R-matrix method. Of these, the hyperspherical treatment, which singles out the total electronic moment of inertia as an adiabatic coordinate, has been more valuable in providing a visual picture of the global evolution of electron correlations in energy and radius. The R-matrix method has evolved instead into a simple, efficient, and accurate procedure for quantitative calculations of photoionization and scattering processes. To date, however, the applications of this finite-volume variational method have been restricted primarily to the lower-lying doubly-excited channels of any given atom or molecule.

Hyperspherical studies have recently been extended to a wide variety of physical systems, including the H_2^+ and HD^+ molecules, the positronium negative ion, and also including various triatomic molecules within the context of a Born-Oppenheimer approximation. These studies have clarified the validity of an adiabatic treatment of the moment-of-inertia coordinate, while also sharpening our understanding of its major shortcomings: the poor convergence of the adiabatic expansion at large distances and at high

energies. Nevertheless, calculations of the potential curves for these systems and for "few-electron" atoms (He, Be, Mg, Ca, Li) have clearly shown that the strongest lower-lying multiply-excited states are predominantly reached by following one or two hyperspherical pathways which are well localized in configuration space.

Two developments within the past year have suggested that the task of constructing potential curves for more interesting open-shell atoms is now within our grasp. The first key has been the introduction of an elegant fractional parentage expansion[1] for constructing a complete set of antisymmetrized harmonics for any number of atomic electrons. The second has been the identification, through a "prediagonalization", of a small subset of these harmonics which dominate the Fourier expansion of the adiabatic hyperspherical wavefunction.[2]

One spectral feature which has come into increasingly sharp focus is the remarkable regularity of atomic spectra along a row of the periodic table. This has emerged both from experiment[3] and from R-matrix calculations[4]. The current status of our understanding of this property will be reviewed at the meeting.

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Doubly Excited States

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Doubly excited atoms, with two excited electrons, occur both above and below the first ionization limit. The interactions between the doubly excited states and the energetically similar singly excited channels are manifested by autoionization and series perturbations in these two cases. Historically these states have been studied by ultraviolet photo-absorption from the ground state. The absorption of the doubly excited state can be thought of as originating in the small excited state admixture of the ground state. This absorption is neccessarily weak with the result that it is comparable to direct photoionization. As both the excitation to the autoionizing state and the excitation to the continuum lead to an ion and an electron these two processes interfere to produce the familiar Beutler Fano profile characteristic of autoionizing states.

Using laser techniques it has become possible to drive the transitions of each of the two electrons separately due to the fact that several photons can be used. This can be done either implicitly in true multiphoton excitation or explicitly using stepwise excitation of real levels. The latter affords one substantially better control over the excitation process.

The basis of the stepwise excitation technique is easily understood by considering Ba as an example. If Ba is excited from its ground state $6s^2$ to a bound $6sn\ell$ state by lasers which are set to fixed wavelengths the $6sn\ell$ state serves the starting point for the excitation of the doubly excited $6pn\ell$ states. The laser which drives the $6sn\ell \rightarrow 6pn\ell$ transition is scanned in wavelength across the $6sn\ell \rightarrow 6pn\ell$ transition which is located near the Ba^+ 6s - 6p transition. In most cases the observed lineshape is, to a good approximation, a Lorentzian giving the immediately the position and width of the autoionizing state. In the language of conventional uv spectroscopy this corresponds to an infinite Fano q parameter, which occurs when there is no continuum excitation. Why there is no continuum excitation is clear from a simple physical picture of the excitation. In

the 6snℓ atom we drive the Ba^+ 6s $\rightarrow$ 6p transition and the outer electron is a spectator. Thus the large oscillator strength of the Ba^+ resonance line is spread over the few wavenumbers corresponding to the width of the 6pnℓ state. The direct continuum absorption of the nℓ electron is much less likely due to the gross mismatch between the spatial oscillations of the wave functions of an nℓ Rydberg electron and a 2eV continuum electron at the classical turning point where the Rydberg electron is likely to be found. For all practical purposes the excitation is only to the 6pnℓ state, not to the continuum.

Applying this excitation scheme to atoms in a beam allows one to probe many facets of the doubly excited autoionizing states. First one can measure the total autoionization rates (radiative decay is asumed to be negligible by comparison) and energies of the autoinoizing states. For simple autoionizing series the multistep method has no great advantage over conventional uv spectroscopy, but for cases in which there are interacting series the clarity of this method has allowed the spectra to be untangled for the first time.

The use of an atomic beam allows the use of more sophisticated techniques than can be employed in absorption spectroscopy. Two good examples are the detection of photons from excited ions and the ejected electrons. Measurement of the electron energy and angular distributions provides much more detail than total ionization rates since the final state of the ion and electron can be better determined, allowing more stringent comparison with theory. Perhaps more interesting though is the fact that the electron spectroscopy reveals phenomena that are masked in the total autoionization rate measurements. Examples are the variation of asymmetry parameters a cross perturbed levels and along perturbed series. Neither of these occurs for unperturbed autoionizing series. Similarly the inhibition of autoionization to specific outgoing channels can be observed when detecting electrons, a phenomenon which is masked in the ion spectrum. The detection of the photons from the excited ions produced is essentially the complement of the detection of energetic electrons and provides the corresponding information.

This work has been supported by the Department of Energy, office of Basic Energy Sciences.

A Pair Description of Two-Electron States

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Two electron states may be divided into two classes. Those in which only one electron is excited, or in which when both are excited they have very different degrees of excitation, form a group distinct from those in which both electrons share comparable amounts of excitation. Particularly for the latter, an appropriate description is one which views the pair of electrons as a single entity from the start. Such states have been called "ridge states" and may be distinguished from the other group of states with disparate excitation by using the term "planetary states" for the latter. In this contribution, we take further steps in the evolving description of a basis for two-electron states in terms of the pair. Thereby we have a complementary description and basis to the independent particle picture.

Whereas conserved quantities like H, ℓ^2 and ℓ_z provide a complete labeling for one-electron states, the corresponding ones for two-electron states H, L^2, L_z and π (the parity operator π has to be independently specified unlike in the one-electron case), where these are the operators for the full system, do not give a complete specification. In the independent particle picture the extra labels to make the specification unique come from the independent coordinates r_1, r_2, $\hat{r}_1$ and $\hat{r}_2$ and the corresponding quantum numbers N, n, ℓ_1 and ℓ_2 (we assume $N<n$). Because of the electron-electron interaction these are, however, not conserved and the more

correlation there is in the system the more is the mixing of values of N, n, ℓ_1 and ℓ_2. In the pair description, two other quantum numbers K and T, which were initially developed in a group theoretic scheme, replace ℓ_1 and ℓ_2. They may be called angular correlation quantum numbers and are relevant for both planetary and ridge states which have strong correlations of this kind. Another quantum number A, taking on the values ±1 or 0, has been introduced to denote certain aspects of the radial correlations. In particular, ridge states must have A = 1 and all A = 0 states are planetary states. We will show that the enumeration and classification of all doubly excited series converging to a single electron N limit can be carried out purely in terms of pair quantum numbers like L, π, K and T (and S, the total spin, if spin aspects are included), removing any necessity to look back at ℓ_1 or ℓ_2.

For ridge states N and n also are more appropriately replaced by pair quantum numbers. In particular, a pair principal quantum number ν to go with the overall "radius" $R = (r_1^2 + r_2^2)^{1/2}$ of the system, together with a Rydberg formula appropriate to a Coulomb potential in R, organize the two-electron states. As with single electron states in any atom other than hydrogen, the Rydberg formula involves parameters such as the quantum defect with a dependence on the other quantum numbers - in our case on K, T, L, S and π. Sequences of numerically available data on doubly excited states have been fitted to the Rydberg formula to extract these dependences and will be presented. Also a direct calculation of the two-electron Hamiltonian appropriate to ridge states, namely $\vec{r}_1 \approx -\vec{r}_2$, will be presented to show a sequence of energy levels with precisely the structure of the Rydberg expression involving ν. These are all elements to understand further the pair description and pair basis for two-electron states.

NOTES

TUESDAY, OCTOBER 21, 1986

MERCER III
10:15 AM–11:35 AM

II-TuA4–7

TWO ELECTRON SYSTEMS

Michele Crance, Laboratoire Aime Cotton, France,
Presider

II-TuA4-1

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Doubly excited states have been in the focus of experimental and theoretical research of atomic and molecular physics for a number of decades. An outstanding problem in this field is the so-called Wannier problem (1-3). The related progress has not yet been able to provide rigorous quantum mechanical, many-electron explanations and the concepts and information currently employed are either classical or semiquantitative.

In this report we present a theory for the a priori identification and accurate calculation of a particular class of doubly excited states which lead-as a function of excitation energy-to the "Wannier state" at the two-electron ionisation threshold of any atom in any symmetry (4). The theory is state-specific (5) and uses suitably defined orthogonal MCHF zeroth-order wavefunctions which correspond to the state of lowest energy in each manifold defined by a particular symmetry in each shell. The choice of the zero-order wavefunctions is systematic and allows for the self-consistent computation of the most important angular and radial (i.e beyond the hydrogenic Hamiltonian) correlations. The remaining electron correlation is added variationally subject to specific orthogonality constraints.

Our first application is to the $^1P^0$ Wannier two-electron ionisation ladder(TEIL) in H^-, He and Li^+. We offer a conceptualization of the theory in terms of conditional probability density plots which show how the localisation around $\langle r_1 \rangle = \langle r_2 \rangle$ and $\langle \theta_{12} \rangle = \pi$ increases as a function of the principal number n

(we went up to n=10). As the excitation energy increases, the significance of the radial correlation beyond that included in the MCHF zeroth-order vector is reduced to negligible levels. The predicted energies and corresponding oscillator strenghts should be observable in photoabsorption experiments. The energies are fitted to an effective Rydberg-like formula $E_n = -(Z-\sigma)^2/(n+\mu)^2$. For H^-, $\sigma=0.161$, $\mu=0.354$, for He $\sigma=0.162$, $\mu=0.160$ and for Li^+ $\sigma=0.168$, $\mu=0.101$. Information from the wave-functions is used to derive the angular correlation of the two electrons emitted at threshold. It is hoped that when numerical accuracy permits it, the appropriate extrapolation of the two-electron photoexcitation cross section to threshold will allow the derivation of a reliable two-electron ionisation law.

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Regularities of Negative Ion Resonances

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The effects of electron correlation in atomic and molecular systems are perhaps most pronounced in negative ions. No long-range Coulomb field is present to bind an additional electron to a parent state of a neutral atom. The stability of a negative ion state is thus usually attributed to one of two alternative mechanisms, both of which invoke electron correlation phenomena: i) polarization of the parent atom provides an attractive potential in which an electron can be weakly bound; ii) two electrons orbit a singly-charged positive ion in a highly correlated fashion, so that each electron is effectively subject to the Coulomb potential of a fractional charge. These correspond respectively to motions in the valleys and upon the ridge of a hyperspherical potential surface. Both mechanisms seem capable of generating the long series of resonances converging on the parent ionization limit, which have been observed in most negative ion systems.[1]

We are attempting to identify the relative importance of these mechanisms by studying elemental sequences. An example of our approach is given in Fig. 1, which shows calculated[2,3,4] eigenphases for electron scattering by Ne, Na, and Mg in an energy range containing the "b" resonance of Ne and the threshold shape resonances of Na and Mg. Eigenchannel analysis was used[2] to identify the "b" resonance as involving definite LS coupling of incident and excited 3s electrons as a $^3P^o$ pair, which would be only weakly coupled to the residual $2p^5$ core. This is characteristic of a ridge resonance. Comparison with Na substantiates the LS identification, since the Na coupling scheme is unambiguous. However, the Na resonance is probably a valley state, consisting of a p wave weakly trapped in an extended ($r \sim 20\ a_0$) barrier structure formed by centrifugal and polarization potentials.[5] Furthermore, in Mg we see a resonance which looks very

similar, even though the parent state contains two rather than one 3s electrons. It seems reasonable to identify these three states as members of the same class. Thus, definite LS coupling of an excited electron pair does not necessarily imply predominance of ridge behavior.

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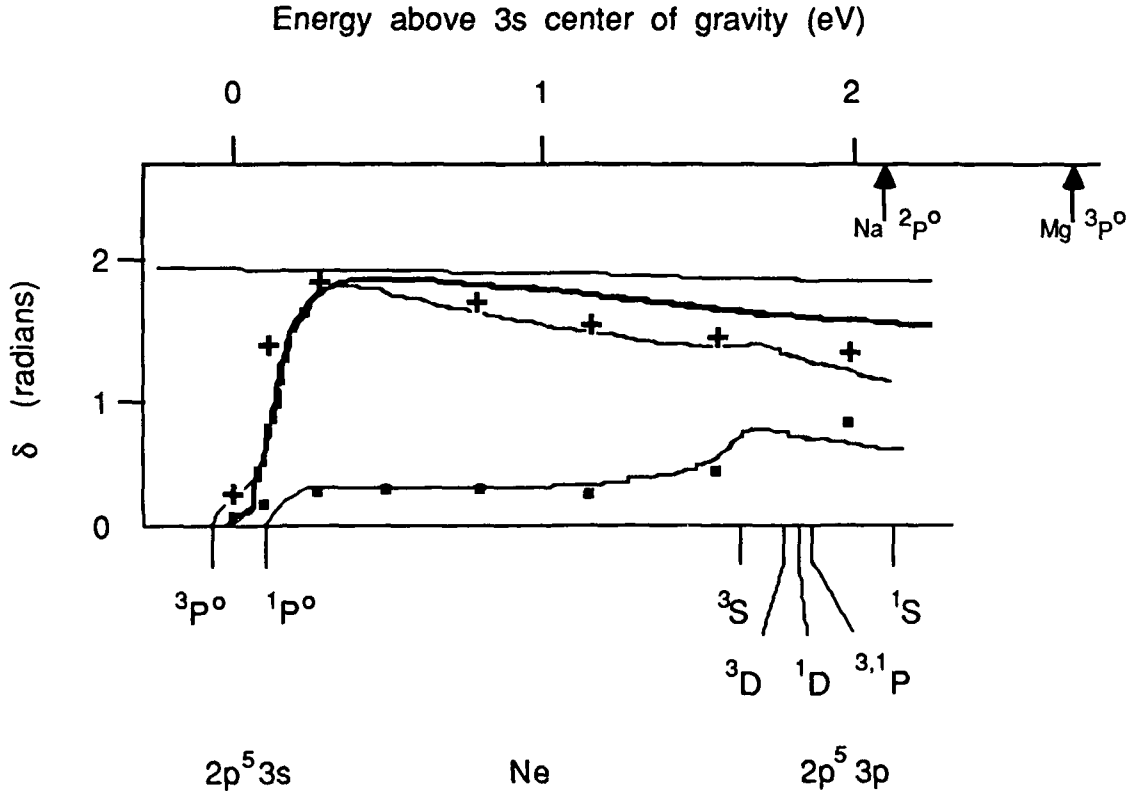
Figure 1. Eigenphases for electron scattering by Ne, Na, and Mg, as a function of energy with respect to the center of gravity of the 3s configuration (ground states of Na and Mg; first excited configuration of Ne). Thin lines, from ref. 2: Eigenphases for e + Ne, ^2S symmetry, open channels being $2p^6\varepsilon s$, $2p^5 3s(^3P^o)\varepsilon' p$, and $2p^5 3s(^1P^o)\varepsilon'' p$. Crosses and points, from ref. 3: e + Na, respectively $^3P^o$ and $^1P^o$ symmetry, open channels being $2p^6 3s\varepsilon p(^3P^o)$ and $2p^6 3s\varepsilon p(^1P^o)$. Bold line, from ref.4: e + Mg, $^2P^o$ symmetry, open channel being $2p^6 3s^2\varepsilon p$. Energies of states in the 3p configurations are indicated.



Novel motions in highly excited two-electron atoms

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Novel attracting motions consisting of periodic, quasiperiodic and chaotic components predict a new resonance formation in doubly excited atoms. Also double escape correlations are analysed.

Inhibited autoionization in multi continua systems

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The phenomenon of "inhibited autoionization",[1,2] which has recently received considerable attention, occurs whenever the direct autoionization amplitude of a given Rydberg series is cancelled by interference with indirect autoionization through a neighboring perturber state. Inhibited autoionization thus requires at least a three channel system, one open and two closed channels, where it was first observed as dramatic reduction of the linewidth of the autoionizing Rydberg series in the vicinity of the perturber [2]

At high excitation energies autoionizing Rydberg atoms are very unlikely approximate three channel systems, therefore we have experimentally and theoretically studied the phenomenon of inhibited autoionization in a more general multi channel case. Specifically, we have investigated the decay of the Barium $[6p_{1/2}ns]_{J=1}$ autoionizing Rydberg series (labeled as channel 4 in the following) into any of the three continua $Ba^+[6s_{1/2}]$ (channel 1), $Ba^+[5d_{3/2}]$ (channel 2), and $Ba^+[5d_{5/2}]$ (channel 3). The autoionizing series is perturbed by two other closed channels, $Ba[6p_{3/2}ns]_{J=1}$ (channel 5) and $Ba[6p_{3/2}nd]_{J=1}$ (channel 6), respectively. Theoretical analysis of the problem, following the lines of ref. 1, yields the total normalized linewidth $\Gamma\nu_4^3$ of the autoionizing series as the sum of the three partial equivalent widths $\Gamma_i\nu_4^3$ into the three continua i=1,2, and 3. Each Γ_i is given by

$$\Gamma_i = (2/\pi)\, Z_i^2/N$$

with

$$\begin{aligned} Z_i = {} & [-R_{i4}R_{56}^2 + R_{i6}R_{45}R_{56} + R_{i5}R_{46}R_{56}] + \\ & + R_{i4}\tan\pi(\nu+\delta_5)\tan\pi(\nu+\delta_6) - R_{i5}R_{45}\tan\pi(\nu+\delta_6) \\ & - R_{i6}R_{46}\tan\pi(\nu+\delta_5). \end{aligned}$$

N is a function of the interaction matrix elements R_{jk} and phase shifts δ_k similar to Z_i, but common to all decay channels, and $\nu = \nu_5 = \nu_6$. The numerators Z_i are quadratic in the energy parameter $\tan \pi\nu$, thus allowing for either two, one, or no real zero in any of the partial decay rates. Even

if zeros (inhibited autoionization) occur in more than one continuum they do, in general, not coincide in energy, making the total linewidth insensitive to the phenomenon. The branching ratio, on the other hand, and, hence, the population distribution of the final Ba^+ ions is strongly affected.

Experimentally, we have used high resolution time-of-flight spectroscopy of the autoionization electrons to observe the partial decay rates into the three continua. As demonstrated in fig. 1 inhibited autoionization has been found in the decay of the $[6p_{1/2}ns]$ - states into the $Ba^+[5d_{5/2}]$ continuum around n = 27 ($\nu_{mod1} \simeq 0.6$, whereby preliminary numerical fitting suggests another narrow zero around $\nu_{mod1} = 0.8$). By comparison of the three decay rates at various energies we found that the population distribution of ions can very effectively be varied through the proper choice of the autoionizing state through which the ions are created. In principle, inhibited autoionization may lead to complete population inversion in multi continua systems, which appears as very attractive feature in the improvement of autoionization pumped UV - lasers.

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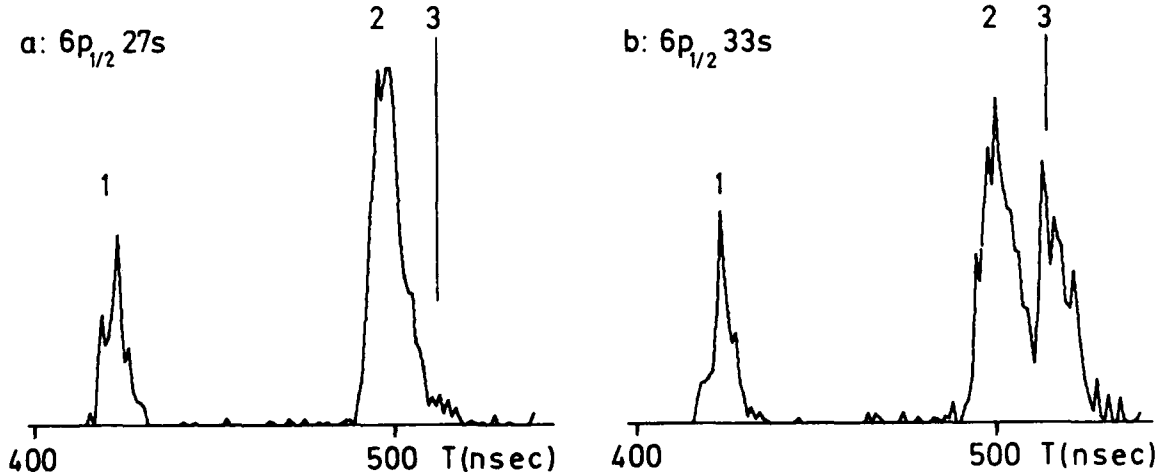

Fig.1 a: Time-of-flight spectrum showing the relative autoionization rates of the $Ba[6p_{1/2}27s]$ state into the continua 1,2, and 3, with inhibited autoionization occuring in continuum 3. b: "Regular" autoionization electron spectrum, shown for comparison ($Ba[6p_{1/2}33s]$ state).

TUESDAY, OCTOBER 21, 1986

MERCER III
1:30 PM–3:00 PM

II-TuB1–3

MULTISTAGE IONIZATION

See Leang Chin, Laval University, *Presider*

Multiply charged ions and energy spectrum of electrons in multiphoton ionization

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The different aspects of the multiphoton ionization of one-electron atoms are now well understood. They can be described by rigourous theoretical models in the framework of perturbation theory. Recently emphasis has been given to two new developments : (1) the production of multiply charged ions from many-electron atoms, (2) the investigation of electron energy spectra produced in multiphoton ionization of atoms.

The interaction between an intense laser pulse and many-electron atoms leads to the removal of several electrons and the production of multiply charged ions. Up to Xe^{6+} ions have been observed at 1064 nm and 10^{14} Wcm^{-2} and up tp Xe^{8+} ions at 193 nm and 10^{16} Wcm^{-2} corresponding to the removal of all the electrons of the external shell. The production of doubly charged ions have been investigated in detail. Two mechanisms have been proposed to explain the formation of doubly charged ions through multiphoton absorption : a "direct" process in which two electrons are simultaneously removed from the atom ; a "sequential" process which consists of successive ionizations of the neutral atom and the singly charged ion. The conditions under which one of these processes dominates are determined mainly by the wavelength and intensity of the laser field. At long laser wavelengths such as 1064 nm, doubly charged ions are produced through the simultaneous removal of two electrons from the neutral atom when the laser intensity is below the saturation intensity I_S, while the stepwise process becomes dominant when the laser intensity is increased beyond the intensity I_S. In contrast, at short wavelengths, double ionizaton always proceeds in a stepwise proces via singly charged ions. It should be pointed out that the ratio of singly to doubly charged ions for laser intensity below I_S never exceeds 1.5×10^{-2} at 1064 nm. This wavelength effect is even more emphasized in He.

The energy spectrum of electrons produced in the multiphoton ionization of rare gases has shown unexpected features. The energy spectrum does not correspond to a single peak as expected from N-photon ionization described by the lowest order perturbation theory. The energy spectrum generally consists of a series of peaks evenly spaced by an amount equal to the photon energy. The number of peaks strongly depends on laser wavelength. For

example, at short wavelengths, the electron energy spectrum consists of a main peak and a very small peak which correspond to the absorption of one additional photon with Xe. In contrast, the energy spectrum of electrons produced at longer wavelength such as 1064 nm consists of a very large number of electron peaks, which correspond to the absorption of a very large number of additional photons, typically 10 to 100 in the 10^{13} - 10^{15} Wcm^{-2} range. Furthermore, the electron energy distribution is very much changed when the laser intensity is increased. This leads to the disappearance of the first peak in Xe at 1064 nm and about 10^{13} Wcm^{-2}, while the first 27 peaks disappear in He at 10^{15} Wcm^{-2}. This is a typical example of high intensity laser processes.

A model will be presented to successfully explain peak disappearance. There are two key points in the physical description of the process : (1) after having absorbed N photons in the bound-state spectrum of the atom, the electron does not emerge in vacuum but in a strong external field. That means that the final energy should not be $E_f = -E_i + N\hbar\omega$ ($\equiv E_o$), but $E_f + \Delta$ where $\Delta = e^2E^2/4m\omega^2$ represents the quiver energy of the particle in the laser field. This additional energy is obtained by absorbing S extra photons by multiphoton semi-inverse Bremsstralhung, so that $E_f + \Delta = E_o + S\hbar\omega$. Therefore $S > \Delta - E_o/\hbar\omega$. The S peaks of the electon spectrum which do not satisfy this condition are thus cancelled out. (2) The second key point is that the electron does not lose nor gain energy when leaving the traveling wave. This is due to the fact that the particle stops oscillating and loses its quiver energy Δ but is accelerated by the so-called ponderomotive force and gains Δ. As a result, the electron energy is $-E_i + (N+S)\hbar\omega$, provided that $S > \Delta - E_o/\hbar\omega$. The disappearance of electron peaks stongly depends on laser frequency. This is in good agreement with experimental results.

This picture is at variance with previous developed model of Muller and Tip which provides a different explanation for the disappearance of the first Xe electron peak at 1064 nm. It is based on laser-induced shift in ionization energy. The linear increase in ionization energy as a function of the laser intensity gradually makes the Nth-order photon absorption energetically impossible, leading to the suppression of the first electron peak of the electron distribution. But this supposed laser-induced increase in the ionization potential of an atom is not at all consistent with previous accurate multiphoton ionization ion measurement, as well as with slopes measurements than with laser-induced ionization energy shift of Kr which shows an ionization energy shift of 86 $\pm$ 6 cm^{-1} by the laser pulse at $2x10^{13}$ Wcm^{-2}. This shift of 10 meV should have negligible consequences on electron energy spectra measurements.

MANY-ELECTRON DYNAMICS OF HEAVY ATOMS IN INTENSE LASER FIELDS. EFFECTS OF SCREENING AND CORRELATION

by

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In this talk we shall discuss the role of collective effects in multiphoton-multielectron ionization of heavy atoms. In recent work /1,2/ we have developed a theoretical description of multiphoton ionization which takes into account effects of many-electron screening and introduces an *effective laser intensity*. This is accomplished by introducing a RPA-type of dynamically screened electron-photon coupling. Since *every* electron-photon interaction operator is screened, our approach includes multiply excited intermediate levels and provides a description of *dynamic non-linear response*.

We shall present results of an application to 2-photon 1-electron ionization of the 5p-shell of xenon in the photon energy region 0.45-1.4 Ry. This includes the ATI region with free-free ionization. We have calculated the non-linear dynamic response using both a local-density (LDA) and a Hartree-Fock basis. In both approaches, screening of the laser field leads to a dramatic change of the ionization cross section, and on the average the two methods give the same results. There is a huge reduction relative to the independent electron approximation, and there are important new contributions associated with the non-linear response (double-excitations). Screening of only the "first" photon (linear response) leads to important differences everywhere and to huge differences in the ATI region.

Finally we plan to discuss briefly different aspects of many-electron effects in direct and stepwise multielectron ionization.

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TIME DEPENDENT HARTREE FOCK THEORY OF MULTIPHOTON IONIZATION

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The theory of multiphoton ionization for single electron atoms (or pseudo-one electron atoms such as sodium and cesium) is relatively well established from the standpoint of the knowing the relevant equations to be solved and interpreting the results. There is still some debate about the above threshold effects, but ionization rates have been calculated for high order processes, and the results agree well with experiment. This is not the case for multi-electron systems. In the limit of laser fields which are very strong compared to the Coulombic interactions, collective motion should be expected. At lower laser intensities, linear response theory or perturbation theory has proven to be adequate to model the absorption processes. In this regime, the inter-electronic forces are stronger than the photon-electron interaction so that the perturbation series is convergent. For the intermediate regime, say $>10^{14}W/cm^2$, these two kinds of interactions should be treated on the same footing. A complete, exact, many body calculation for this problem is not possible so that several approximations have been considered. Independent particle models seem to be the most reasonable, with time dependent Hartree Fock being able to include most of the important physical effects, particularly as the number of electrons increases. The major approximation of this approach is that the state of the system is represented by a single, determinantal wave function for all time. This constraint limits the kinds of information which can be obtained from the calculations to averaged quantities. It has the merit, however, of including the effects of the time evolution of the electronic charge density on the absorption process. The redistribution of absorbed energy between all the system's electrons, and the rate at which this occurs is part and parcel of the calculations. This energy exchange is done self

consistently so that an understanding of the total absorption dynamics is possible. This seems a minimum requirement in order to investigate any collective effects.

We have studied helium using a single, doubly occupied orbital to represent the time evolving state. We calculate ionization rates for 532nm photons (an 11th order process) and compare those to results where one of the electrons is fixed in its initial ground state orbital. The second model eliminates any possible collective effects, but in both cases the active electrons are absorbing energy from the photon field in the presence of the field of the other electron. Our results show that for field strengths up to $10^{15} W/cm^2$ the ionization rates are independent of the freedom of the second electron. This is consistent with experiments which produce only singly ionized helium at these intensities.

The calculations are carried out using a finite difference grid in two of the spatial dimensions and a basis expansion in the third. Assuming the laser is linearly polarized, the problem is symmetric about the axis of polarization so that a limited number of basis functions in the azimuthal coordinate is required. The time propagation of the electronic wave function is performed using the alternating direction, implicit method of Peaceman and Rachford. The time evolution of the initial, ground state wave function is followed through a "slow" turn on of the matter-field interaction. The field reaches its maximum intensity after 10-20 periods and is constant thereafter. The rate of ionization is determined by either projection onto bound states or absorption of flux at the boundaries of the spatial grid. Other physical parameters of interest such as the orbital energies, expectation values of z and ρ, etc. are monitored also. The results of the calculations are expected to provide new insight into the absorption dynamics.

TUESDAY, OCTOBER 21, 1986

FLAG PAVILION

II-TuC1–16

3:00 PM–5:00 PM POSTER PREVIEWS

6:00 PM–8:00 PM POSTER PRESENTATIONS

II-TuC1-1

DYNAMICS IN INTENSE FIELDS - BEYOND THE DIPOLE APPROXIMATION

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We have recently shown in a series of papers [1-7] that coupled equations methods of quantum collision theory can be conveniently used to investigate processes such as direct photodissociation [1-2], Resonance Raman Scattering in weak and strong fields [3-5], and higher order nonlinear spectroscopies [6-7] for diatomics involving several well isolated excited electronic states. This was done using the dressed molecule picture [8-12] of molecule-radiation interaction, wherein photon states are explicitly included into the theoretical description. All these methods are adequate only in the case of well isolated electronic states. There is an urgent need to derive a priori the most efficient representation for general electron-nuclear-radiation field systems such as occurs in strong field laser chemistry. In recent work we have examined this problem in an effort to incorporate as much as possible the electromagnetic field into the dynamics [13-14]. One might surmise that classical approaches should work sufficiently well at the high field intensities described here, and much work has been pursued in that direction. As we have pointed out previously, this involves treating both the molecule and the field classically. For electron-radiation interactions one would prefer a quantum formulation, since electronic states are, as a result of their large excitation energies, true quantum states. Furthermore, quantum mechanics leads to a linear theory of interactions whereas classical mechanics is a highly nonlinear theory [15]. Thus using a quantum formulation of matter- field interactions [16-17], we have been able to exploit methods of early (non-covariant) quantum electrodynamics (QED). In particular we have shown that the Bloch-Nordsieck (BN) representation [18-20] (which leads naturally to the concept of coherent states in momentum space) was very convenient as a method of introducing strong field effects directly into the quantum dynamics of molecular systems. Coulomb gauge ($\vec{A}.\vec{p}$) and Electric Field Gauge ($\vec{E}.\vec{r}$) representations were shown to be poor zeroth order approximations for the dressed molecular eigenstates in the presence of strong fields, i.e., the adiabatic coupled equations for the latter two gauges define the appropriate adiabatic state as unperturbed (zero field) molecular states [13- 14].

In the present work, we continue to explore the application of the Bloch-Nordsieck representation to intense field-molecule interactions. In particular we shall examine corrections beyond the dipole approximation. Although normally higher order contributions such as electric quadrupole and magnetic dipole interactions are proportional to $(kr)^n$ where n is the order of the multipole, $k = 2\pi\lambda^{-1}$ (photon), so that $kr \ll 1$ for infrared and visible radiation, strong fields impart an oscillatory motion to the electron which can invalidate the dipole approximation . Thus for the case of a circularly polarised incident wave of amplitude E, the electron follows the field in a circle with angular velocity ω equal to that of the wave, so that $eE = m\omega^2 r = m\omega v$. This gives $v/c = eE/m\omega c = \frac{eE/k}{mc^2}$. eE/k is essentially the potential difference across one wavelength and mc^2 is the electron rest mass energy. Comparing with kr, we obtain $kr = \left(\frac{\omega}{c}\right)\left(\frac{eE}{m\omega^2}\right) = v/c$. This simple exercise shows us that field induced distortions of electron orbits can render the dipole approximation <u>inoperative</u> at high fields, especially for low frequencies which give rise to field induced tunnelling [21]. As an example, for $v/c \simeq 1$, one obtains $E \simeq 3 \times 10^6$ volts/λ, so that for a CO_2 laser ($\lambda = 1000\ cm^{-1}$), $E \simeq 3 \times 10^9$ V/cm which is essentially the hydrogen ground state Coulomb atomic potential. We thus conclude that for field intensities beyond 10^{10} W/cm^2, one has to worry simultaneously about nonperturbative interactions of electronic states as well as corrections to the dipole approximation. It is the purpose of this work to focus on extending the Bloch Nordsieck (BN) approach beyond the dipole approximation and compare it to Electric Field (EF) formulations. Finally it is clear from the above discussion, that when multipole contributions become important, i.e. v/c approaches unity, one should also, in principle, consider relativistic corrections. This question will be addressed in future work on this subject.

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Dressing to the Hydrogen Atom in an Extremely Intense Laser Field

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The mutliphoton ionization rate of atoms in intense laser field is profoundly affected by the "dressing" of the states of the atom by the field. For example, the continuum threshold is known to be shifted upward by the ponderomotive potential. However, little has been done on the dressing of the initial (ground) state beyond lowest order perturbation theory. This is insufficient for intense lasers and this dressing is at least as important to the process as the shift mentioned above.

We have rederived an equation for the dressed bound state of the H atom. It can be interpreted as an H atom in a dielectric medium (provided by the laser). One immediate result is that the level shift is the ponderomotive potential plus smaller terms. Therefore, the effect of the shift of the continuum by the ponderomotive potential is canceled in the calculation of the ionization potential. The remaining intensity dependent terms of the ionization potential are smaller, and are being obtained numerically.

The applicability of the theory to other atoms will be discussed.

INVERSE HALF-BREMSSTRAHLUNG IN MULTIPHOTON IONIZATION OF ATOMS IN INTENSE LIGHT BEAMS

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It is shown that absorption of the minimum number of photons needed to ionize an atom in a light beam of moderate intensity is no longer energetically possible when the external field is strong. However, subsequent absorption of a large number of photons by the "emerging" electron in the field of its parent ion can occur via multiphoton inverse "half-Bremsstrahlung" and ionization is possible again. This new theory leads to a simple interpretation of the appearance of a large number of peaks in the electron spectrum together with the disappearance of the low-energy peaks. The roles played by the laser intensity and wavelength are singled out, and quantitative agreement is found between the conclusions of the present theory and existing experimental data. Furthermore, using an argument developped in a recent paper (Phys. Rev. Lett., **54**, 1385 (1985)), it is demonstrated that the ponderomotive force does not contribute to any modification of the energy of the electron when it leaves the laser beam. An intuitive physical explanation of this fact is given.

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With the goal in mind to separate final state effects due to the interaction of the ejected electron with the laser field from dynamical effects associated with the process of ionization, we investigate the following simple model: We assume an effective interaction that just lifts the electron into the continuum via absorption of the minimum number N of photons which is necessary to overcome the ionization potential (defined in the absence of the field). The effective interaction is left unspecified except its matrix element is assumed to be proportional to $I^{N/2}$ with I the intensity of the laser field. As soon as the electron is free we assume it only feels the laser field and, consequently, is described by the so-called Volkov solution which provides an exact solution for an electron in an external plane wave field. For simplicity, we also adopt the long wavelength approximation for the laser field. For the initial atom and the final ion we take the unperturbed wave functions and we also disregard the recoil imparted to the ion. Altogether, this is essentially the Keldysh approximation. It turns out that boundary conditions, i.e. the way the electrons leaves the laser pulse, are of vital importance.[1] In order to explain the experimentally observed absence of intensity-dependent shifts in the electron spectra we have to assume that the electron leaves the electron pulse on the side rather than being passed over by the pulse. Under these conditions, we also obtain the total suppression of the

low-lying peaks of the electron distribution with increasing intensity which is impressively born out by recent experiments.[2,3] The mechanism can be ascribed to the ponderomotive potential which adds to the ionization potential. When the electron leaves the pulse on one side the energy corresponding to the ponderomotive potential is converted into kinetic energy.[4]

The heights of the peaks of the resulting electron spectra are given in terms of ordinary or generalized Bessel functions for a circularly or linearly polarized laser field, respectively. The calculated spectra exhibit all the universal features of the experimental data aside from the widths of the latter. In view of the simplicity of the model it is not surprising that quantitative agreement with the data[2] could not be achieved, in general. At higher intensities significant differences between linear and circular polarization begin to show up.

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Strong Field Laser Ionization of Model Atoms

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Recent experiments on the ionization of atoms under strong field lasers has renewed interest in the theoretical modeling and calculation of such nonperturbative processes. A promising calculational method is the direct solution of the time-dependent Schrodinger equation using numerical techniques developed to solve parabolic partial differential equations on vector processing computers. We are currently working on numerical solutions to the one-dimensional equation (in atomic units):

$$i \frac{\partial \psi(r,t)}{\partial t} = -\frac{1}{2} \frac{\partial^2 \psi(r,t)}{\partial r^2} + V(r,t)\ \psi(r,t) ,$$

where V(r,t) is a finite square well potential subject to an arbitrarily strong electromagnetic field. We have used the Galerkin method with linear tent finite elements to check the one bound state results of Goldberg and Shore[1] and the two bound state results of Austin[2]. We plan to use the Galerkin or collocation method with B-spline finite elements to investigate the one-dimensional Coulomb problem. The most favorable numerical method will be extended to two or more dimensions.

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General Relativistic Ponderomotive Force in a Moving Medium

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ponderomotive force is an important factor in intense field electrodynamics and plays a fundamental role in the ultrapowerful laser interaction with free electrons, atoms and plasma and in determining the electron energy spectrum in multiple- photon ionization. Therfore, the study of the ponderomotive force and its various effects has stimulated people's great interest.

Nonrelativistic and relativistic forms of ponderomotive force have been derived[1], but these results are based on a single particle model and take no account of the collective effect of surrounding particles. Obviously, they are not quite suitable to the media such as plasma and solid material.

In this paper, the first general covariant derivation of the ponderomotive force is given, by using the concept of "optic metric" $\bar{g}_{ab} = g_{ab} + (1 - \frac{1}{\varepsilon\mu})v_a v_b$[2], in the frame of the general theory of relativity, in which the "optic metric" is equivalent to the "collective effect". For an arbitrarily polarized light with frequency ω, wave number k and four-vector

$$A_4 = A_1 = 0$$

$$A_2 = \frac{c}{\omega} a_1 E_0(x)\sin(kx - \omega t)$$

$$A_3 = -\frac{c}{\omega} a_2 E_0(x)\cos(kx - \omega t + \delta)$$

we obtain the expression for the general relativistic ponderomotive force in the medium moving with constant velocity

$$F_{pd} = (\varepsilon\mu)^{1/2} n f_{eff} = (\varepsilon\mu)^{1/2} n\left[-\frac{\omega}{2\pi}\int_0^{2\pi/\omega} dt\ \frac{mc^2}{V^{(4)}} X\right]$$

Here X and $V^{(4)}$, similar to X and γ in Ref./1/, depend on the "optic metric" apart from the light field. The discussion of several special cases has shown that the effective non-linear force f_{eff} acting on a single particle in static medium is different from that in vacuum. The previous derived expression is valid in some range only if the ponderomotive force F_{pd} is considered acting on the particles in unit proper volume. For relativistic case, our treatment of a single particle has involved the motion due to the force F_{pd} itself, and then the result is more exact than Lindman et al's/1/.

For the medium moving with constant velocity v_x and irradiated by circularly polarized light, we arrive at a new conclusion that F_{pd} is g^{11} times as large as one in static medium or vacuum. Here α is related to the "optic metric" and light field, and $\alpha \longrightarrow$ as $\varepsilon \longrightarrow 0$. This means that the ponderomotive force can become unlimited for the moving plasma with the critical density.

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CLASSICAL CALCULATIONS OF THE EFFECTS OF HIGH BRIGHTNESS LASER RADIATION ON PHOTOELECTRON ANGULAR DISTRIBUTIONS

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A two step model has been developed to describe the effects of a high brightness laser on the angular distributions of photoelectrons near and above threshold. The photoelectron production process and the transport of the electrons away from the ion are decoupled. The model predicts that the photoelectron distribution becomes more isotropic as the laser intensity increases. A larger laser intensity is required to perturb higher energy photoelectrons.

At laser intensities strong enough to produce significant multiphoton ionization, the photoelectron angular distribution is strongly peaked along the direction of the laser electric field for linerarly polarized light (1). At higher laser intensities the electromagnetic field of the laser will perturb the photoelectron orbit in the region of the ion resulting in a trajectory that is dramatically different from the low intensity case. The mode separates the production of the photoelectrons from their transport away from the ion, i.e. a two step process.

The photoelectron is given a velocity (energy and direction) in the region of the ion to give it a direction consistent with low intensity photoelectron distributions, i.e. along the laser electric field direction. The energy of the photoelectron at a large distance (infinity) is selected to be consistent with threshold or above threshold processes. This models the photoionization process.

The electron orbit is calculated on a computer using a parametric relativistic rescription developed by P. H. Y. Lee (2). The laser is described as a traveling electromagnetic wave. The ionic field is a central potential. The photoelectron is assumed to be emitted at the peak of electromagnetic wave. The electron trajectory can be calculated with laser intensity and photoelectron energy and direction near the ion as parameters. By comparing low intensity and high intensity trajectories for the same initial conditions, significant differences can be seen in the photoelectron angular distributions.

Two conclusions may be drawn from the results of these calculations. One, as the laser intensity increases the angular distribution becomes more isotropic. Two, the higher the electron energy a larger laser intensity is required to perturb the distribution. These conclusions indicate that threshold ionization electrons will be perturbed more than above threshold ionization (higher energy) electrons.

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Model of Two-Electron Ionization Based on Semi-Classical Phase Space Averaging

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In this work the two electron ionization of helium is studied with a mixture of quantum and classsical theories.

Prior to laser excitation the state of the helium atom is described by the ground state wavefunction which to a good approximation is equal to $Z^3/8\pi \exp\left[-Z \left(|\vec{r}_1| + |\vec{r}_2|\right)\right]$, where $Z = 1.69$ and $\vec{r}_1$ and $\vec{r}_2$ are coordinates of the electrons. The connection between the quantum-mechanical wavefunction and the classical concepts is provided by the Wigner function. The coarse-grained Wigner function is interpreted as a classical probability distribution in the phase space. A representative sample of 300 points in the phase space is chosen with the help of Monte-Carlo "importance" sampling. This sample represents the initial state of the atom.

The interaction of the atom with the laser pulse is described by the Newton equation of motion which take into account the Coulomb forces between all charges and forces exerted on the electrons by the external laser field. These equations are solved numerically for several field

strengths and frequencies. The initial conditions are provided by the representative sample described above.

The state of the atom is analyzed for times up to 360 atomic units. The following criterion for ionization was adopted. An electron is said to be ionized if its compensated energy $(p - A/c)^2/2 - 2/|r|$ is positive and its distance from the nucleus $|r|$ is larger than some fixed radius r_0, usually taken to be equal to 32 atomic units. According to this criterion the single and double ionization probabilities are found. For example, for the laser field strength E = 1 atomic unit (corresponding intensity 10^{17} W/cm^2) and frequency 0.5 atomic unit (wavelength ~ 100 nm) the ratio of double to single ionization probabilities reaches 0.55 at t ~ 200 atomic units (~16 optical periods) and remains nearly constant for times up to 360 atomic units.

In summary, two striking features found in this model are:

(1) A steady state is reached without complete double ionization with a substantial number of singly ionized atoms remaining.

(2) There is a very small contribution of the stepwise ionization process as compared to the direct two-electron process. Stepwise ionization can be seen only for field strengths larger than 1 atomic unit. The lack of any existing fully quantum treatment of ionization at such field strengths (corresponding to intensities above 10^{17}W/cm^2) is one of the attractions of our semi-classical approach.

II-TuC9-1

Collective Dipole Exitation of Many-electron Atoms by Intense Laser Pulse

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A major implication of the remarkable data[1-3] on multiple ionization of many electron atoms in intense laser pulses is the unusually strong coupling strength at UV and infrared frequencies. Luk et al.[2] appear to have been the first to envisage a giant dipole or multipole excitation of atoms, in the present context. The purpose of this paper is to give a many-body non-perturbative analysis (using a generalization of a method of Tomonaga[4] within the random phase approximation) of an N-electron atomic Hamiltonian coupled to a laser pulse and to demonstrate the existence of laser driven colletive dipole excitations of noble gas atoms. We also determine the photon absorption cross-sections due to this process. It is shown that the dipole operator of a many-electron atom can execute quantum oscillations with a distribution of fundamental frequencies and that the laser excites preferentially the mode whose frequency is resonant with the laser frequency. These modes appear essentially in the outer region of the atomic charge distribution, both at 193 nm and 1064 nm laser wavelengths. It is found, for example, for Xe that the $5p^6$ and the $5s^2$ sub-shells alone contribute virtually completely to the resonant modes; all the other shells contributing less than 0.1% .

Table 1 below summarises the numerical results for the cross-sections of photon absorption due to collective dipole excitation at 193 nm and 1064 nm in several noble gas atoms.

Table 1. Absorption cross-section σ_{ab} due to collective dipole excitation of noble gas atoms at 193 nm and 1064 nm. Also shown is the mean energy $\langle\Delta E\rangle$ absorbed by an atom, at an intensity $I=10^{14}$ Wcm^{-2}, within an assumed effective interaction time of 50 fsec.

Atom	193 nm		1064 nm	
	$\sigma_{ab}(a_0^2)$	$\langle\Delta E\rangle$(eV)	$\sigma_{ab}(a_0^2)$	$\langle\Delta E\rangle$(eV)
Xe	8.4	319	16.1	111
Kr	7.0	265	13.6	94
Ar	5.7	214	11.5	80
Ne	2.4	98	4.8	33

To conclude: laser driven collective dipole excitation is shown to provide a strong and rapid mechanism with large cross-sections for absorption of laser photons by noble gas atoms.

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II-TuC10-1

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Measurements of the energy distributions, Branching ratios, and angular distributions of electrons ejected from SrI $5p_{3/2}ns_{1/2}$ and $5p_{1/2}ns_{1/2}$ J=1 doubly excited states with n ranging from 10 to 20 in the autoionization to the SrII 5s, 4d, and 5p ion state have been made by the techniques of Isolated-Core-Excitation and Time-of-Flight Spectroscopy. The experiment consists of three Nd:YAG laser pumped tunable dye lasers which excite the Sr atoms in an atomic beam from the ground state to the $5p_{3/2}ns_{1/2}$ or $5p_{1/2}ns_{1/2}$ J=1 autoionizing states. The autoionizing electrons are analyzed in energy and angle by a time of flight analyzer, and the electrons and residual ions are collected by two microchannel plate detectors respectively and sent to the computer through the Boxcar Integrators. The measurements show that the doubly excited SrI $5p_{3/2}ns_{1/2}$ and $5p_{1/2}ns_{1/2}$ J=1 states autoionize predominantly to the excited SrII 4d ion states with a p or f electron. The asymmetry parameter varies continuously within the line profile of the $5p_{3/2}ns_{1/2}$ J=1 states as the manifestation of the localized interaction between the SrI $5p_{3/2}ns_{1/2}$ and $5p_{1/2}ns_{1/2}$ J=1 autoionizing series. This variation is distinguished from that caused by the interference between the excitation to the bound part of the autoionizing state and underlying continua.

The six channel MQDT analysis which is made consistent with the treatment of the total autoionization rates, along with the angular momentum transfer theory is utilized to analyze the data. A reasonable agreement between the theory and the experiment data is achieved. For the SrI $5p_{3/2}ns_{1/2}$ J=1 state lying above SrII $5p_{1/2}$ limit, the calculated asymmetry parameters are constant everywhere as the consequence of the absence of the channel interaction.

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Photoelectron Spectrum Resulted From Autoionizing State Resonance

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Autoionizing state resonance is very likely to occur during the above-threshold ionizations of atoms. This paper presents our theoretical studies of the photoelectron spectrum produced by such resonance. By employing the technique of resolvent operator, we have shown that in general there are four peaks in the spectrum (as shown in Fig.1), two of which result from the autoionization of the autoionizing state,[1] others are related to the photoionization of the autoionizing state. Their relative positions and widths are determined by the following expressions:

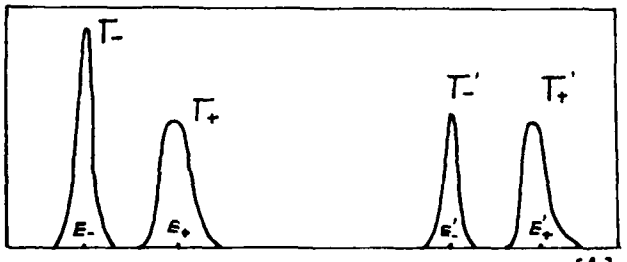

Fig.1 Schematic diagram of the photoelectron spectrum

$$E_{\pm} = -\frac{1}{2}\Delta \pm \frac{1}{2}\left[\Omega^2 + \Delta^2 + \gamma_a\gamma_b - \frac{(\Gamma_a + \gamma_a + \gamma_b)^2}{4}\right]^{1/2}$$

$$E'_{\pm} = E_{\pm} + \hbar\omega$$

$$\Gamma_{\pm} = \Gamma'_{\pm} = \frac{1}{2}(\Gamma_a + \gamma_a + \gamma_b) \pm \frac{\Omega^2/2\beta}{\left[\Omega^2 + \Delta^2 + \gamma_a\gamma_b - \frac{(\Gamma_a + \gamma_a + \gamma_b)^2}{4}\right]^{1/2}}$$

where Ω is the Rabi frequency, Γ_a is autoionization width, β is

Fano parameter, ω is photon frequency, and γ_a, γ_b are the laser induced stark shifts of the autoionizing state and the ground state, respectively.

From the above expressions, we have obtained the condition on which two of the four peaks(Γ_+, Γ_+' or Γ_-, Γ_-')may be disappeared.

$$32\Delta^2 + 8|q|\Delta(\Gamma_a + \gamma_a + \gamma_b) + (\Gamma_a + \gamma_a + \gamma_b)^2(q^2-4) \geq 0$$

We have also derived the critical value Ω_0 of the Rabi frequency at which two of the four peaks are disappeared. In the case of $\Delta = 0$, $q = 5$, we have found that $\Omega_0 \doteq 2\sqrt{6}\,\Gamma_a$, which is identical with that obtained in [1] .

Taking account of the natural width of the autoionizing state does not affect the two peaks near the autoionizing state, but the other two peaks are shown to no longer disappear.

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Multiphoton Isolated Core Excitation

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In a number of recent experiments, the Isolated Core Excitation (ICE)[1] method for producing doubly excited Rydberg states has been extended to two photon core excitations.[2,3] One particularly novel experiment showed that it is possible to use two photon excitation even in the case where the single photon excitation moment is zero.[3] Furthermore, an analytical, closed form expression for this two photon core excitation moment has been reported.[4] This work extends that analysis to the general case, where the core has absorbed n independent photons.

The ICE method consists of exciting a core electron after an outer electron has already been placed in a Rydberg state.[1] Since the Rydberg electron is far removed from the core, the core has effectively been "isolated". The Rydberg electron does have an effect on the transition moment, however, and this shows up primarily in the shake-up spectra, where the Rydberg electron shakes into nearby Rydberg states with the same angular momentum, but different principal quantum numbers. A multiphoton core excitation moment must then properly account for <u>all</u> possible intermediate "shake-up" transitions, including those which are off the energy shell.

We have developed a closed form for n photon core excitation which explicitly shows both contributions: the "prompt" terms in which the Rydberg electron does not shake until all n photons have been absorbed, and the "stepwise" terms which show the Rydberg electron's readjustment after each intermediate photon is absorbed. Several general features become apparent: (1)

it is possible, in general, to set each photon frequency so that only the n photon transition is non-zero, and all lower order transitions are zero; (2) an intermediate shake-up resonance does not always enhance the net transition rate since this route generally interferes destructively with the "prompt" route; and (3) an intermediate shake-up resonance will enhance the n photon transition rate if the Rydberg electron's binding energy change is larger than the autoionization-induced linewidth of the intermediate resonance, otherwise it will decrease the net n photon transition rate.

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II-TuC13-1

Observation and Analysis of the Autoionizing Spectra of Sr

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The Sr atoms are excited stepwise by means of three dye lasers. All three lasers are linearly polarized in the same direction to allow the populations of the $(5p_{\frac{1}{2}}ns)_{J=1}$ or $(5p_{\frac{1}{2}}nd)_{J=1,3}$. By sweeping the wavelength of the third dye laser we are able to record the excitation spectrum of the autoionizing states.

It shows that the quantum defects of $(5p_{\frac{1}{2}}ns)_1$ are nearly constant. It indicates that the $(5p_{\frac{1}{2}}ns)_1$ series of Sr (n=10-26) has less couplings with other series.The single strong peak is observed in the $(5p_{\frac{1}{2}}ns)_1$ series at the $Sr^+5s-5p_{\frac{1}{2}}$ wavelength. And two strong peaks are observed in the $(5p_{\frac{1}{2}}nd)_{1,3}$(n=17-25).These spectra are explained using a quantum-defect theory appoach which shows that the cross section for photoexcitation 6 is proportional to a product of the spectral density of the autoionizing state and the overlap integral from the initial bound Rydberg state.

Table 1. shows the measured values of Sr $(5p_{\frac{1}{2}}ns)_1$ series .

Table 1. The measured values of the energy levels E, Quantum defects δ and the wavelength of the third dye laser λ_3 of Sr $(5p_{\frac{1}{2}}ns)_1$

n	$E(5p_{\frac{1}{2}}ns)_1$ (cm^{-1})	λ_3(Å)	δ
10	67126.1±0.8	4234.8±0.1	3.403
11	67736.8±0.4	4230.1±0.1	3.421
12	68165.0±0.7	4224.4±0.1	3.396
13	68450.1±0.6	4223.6±0.1	3.426
14	68669.7±0.4	4221.2±0.1	3.407
15	68829.3±0.7	4220.4±0.1	3.418
16	68956.7±0.2	4219.1±0.1	3.394
17	69053.2±0.5	4218.9±0.1	3.409
18	69131.7±0.9	4218.5±0.2	3.412
19	69195.9±0.8	4218.2±0.1	3.410
20	69249.8±0.6	4217.7±0.1	3.386
21	69293.4±0.9	4217.6±0.2	3.393
22	69329.8±0.8	4217.6±0.1	3.411
23	69362.0±0.5	4217.3±0.1	3.391
24	69388.1±0.6	4217.4±0.1	3.427
25	69411.6±0.4	4217.3±0.1	3.426
26	69432.6±0.4	4217.1±0.2	3.396

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Multiphoton Ionization of Beryllium

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We have utilized resonance ionization mass spectrometry (RIMS) to investigate the spectroscopy and dynamics of multiphoton absorption in Be. This work is directed towards developing methods for selective laser ionization of the isotope ^{10}Be, a long-lived radionuclide ($\tau_{1/2} = 1.6 \times 10^6$ a) used for geological dating purposes. Since Be is the simplest "two - electron" atom that can be excited by absorption of a few photons at convenient laser wavelengths, these studies may also contribute to understanding the nonlinear interaction of radiation with many-electron systems.

Previous work in our laboratory[1] investigated two- and three-photon ionization of ^{9}Be (the only stable isotope), with attention focused on the spectroscopy of autoionizing states. The spectral width of the laser used then was too large to allow isotopically selective excitation, so we have replaced it with a pulsed dye amplifier (PDA). [2] A schematic of the new RIMS system is given in Fig. 1. Sample preparation involves mixing several micrograms of $BeCl_2$ with a graphite slurry and placing it on a rhenium filament. The concentration ratio ^{10}Be: ^{9}Be: is approximately 10^{-3}. The PDA produces 25mJ pulses of 10 ns duration at 560 nm with a 150 MHz bandwidth, corresponding to $\Delta\nu / \nu \sim 3 \times 10^{-7}$; by comparison, the normal mass isotope shift for the ^{9}Be:^{10}Be pair is $\Delta\nu / \nu = 6 \times 10^{-6}$. The 560 nm radiation is frequency-doubled by a KDP crystal, yielding ~1 mJ pulses at 280 nm.

Figure 2 illustrates the ^{10}Be: ^{9}Be isotope shift in the two-photon $1s^22s^2\ ^1S_0 \rightarrow 1s^22s6s\ ^1S_0$ transition at 71321 cm^{-1}. This preliminary measurement yields an isotope shift of 13.31±0.05 Ghz. The normal mass shift for this transition is 13.04 Ghz, so the specific mass shift (or mass

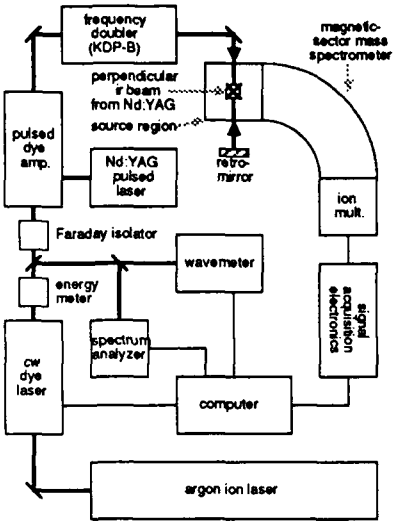

Fig. 1. RIMS II apparatus incorporating pulsed dye amplifier. Counterpropagating UV beams produce Doppler-free excitation of atoms in source region; an orthogonal IR beam photoionizes the excited state.

polarization effect) is a few percent of the total. In the single configuration approximation, the specific mass shifts of the transitions $2s^2\ ^1S_0 \rightarrow 2sns\ ^1S_0$, $2snd\ ^1D_2$ vanish identically; thus their actual values indicate the extent of interaction of the s and d Rydberg series with the doubly - excited $2p^2$ configuration. We shall report results on other members of these Rydberg series at the meeting.

This work is supported in part by the U.S. Department of Energy under contract DEAI05-86ER60446.

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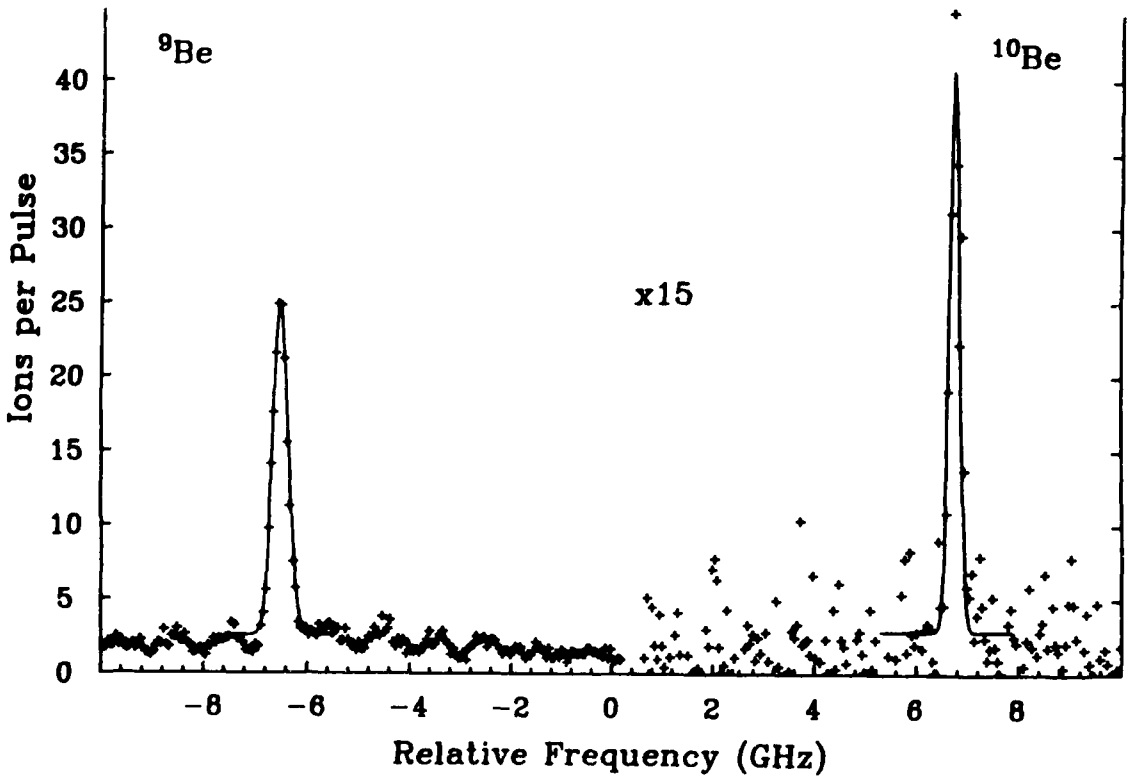

Fig. 2. Yield of ^{9}Be and ^{10}Be ions as a function of laser frequency for the two-photon 2s → 6s transition. Central break reflects change of mass analyzer channel.

Electric Field Effects on Doubly Excited Autoionizing States

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We have observed electric fields to have pronounced effects on autoionizing resonances in the continuum region above the first ionization threshold. These effects are observed predominantly between field-coupled levels of opposite parity that have significantly different zero-field widths. The types of phenomena come under three categories:

1) Field-induced broadening (and narrowing)

We have reported the observation [1] of a Sr resonance to broaden approximately quadratically in an electric field due to its field-mixing with a nearby much broader resonance of opposite parity. We have also observed the width of a narrow high J resonance in Gd to increase stepwise quite suddenly at a field of less than 2 V/cm. We attribute this to the level's lying near a series of Rydberg levels with $n \sim 60$. The sudden increase in width is observed at the Ingliss Teller limit for this n, where static manifolds of different n levels overlap. For this field and higher the Stark manifold spans the energy region, and, due to anticrossing between n-manifolds, J mixing becomes nearly complete. Thus, field mixing of these Rydberg levels with an interloping level, even one of high J, occurs essentially for all fields at and above the Ingliss Teller limit.

2) Field-induced interferences

In an electric field, a relatively narrow resonance that is nearly degenerate with a much broader resonance of opposite parity has been observed to cause an interference dip in the broad level that deepens as the field is increased, until at higher fields the broad level is split into two components.

Also, narrow resonances in the wings of broad resonances lead to field-induced Fano-Bentler type inference patterns whose amplitude increases as the field is increased. Results thus far are in good agreement with a non-perturbative theory developed recently [2].

3) Anisotropic core effects

Rydberg levels with $n \gtrsim 5$ exhibit quasi hydrogenic Stark manifolds in which the energies of the nearly degenerate higher-ℓ levels with small quantum defect "fan out" linearly in an electric field. Elegant studies of this phenomena have been made for bound levels of alkali and alkaline-earth atoms [3]. These cases correspond to a spherically symmetric core. The manifolds are strikingly different when the ionic core (seen by the Rydberg electron) has angular momentum $j > 1/2$, which is the case for most autoionizing levels and for most bound levels except those in the first two rows of the atomic table. The angular momentum ℓ of the Rydberg electron couples electrostatically (via the r_{12}^{-1} interactions and the tensor polarizability of the ion core) with the angular momentum j of the core to give rise to (2j+1) fine structure components (when $\ell > j$). As the field is increased, more and more of these components are observable from a given lower level. The Stark-induced levels appear to split into more levels; hence the term "procreative" Stark manifold. We have performed detailed calculations (in jK coupling) of this phenomenon and compare them with our observations of Stark manifolds in barium autoionizing Rydberg levels.

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II-TuC16-1

Double Electron Excitation of Lithium by Electron Impact

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Excitation of the autoionizing states caused by single inner shell electron excitations in alkalai atoms by electron impact have been well studied using various approximations. These include the Born, Glauber[1] and distorted wave theories.[2] In lithium, in addition to single excitation, there have been some doubly excited states observed. The doubly excited states with the two electrons are the most fundamental atomic species which autoionize. Such doubly excited states in helium and alkaline earths have also been studied.[3-5] A first step towards the study of such excitations in lithium would be to find the cross sections for their production by electrons. The first calculations of this nature were done by Kulander and Dahler[4] using the simple Born Oppenheimer approximation. They reported results for $Li(1s)^2(2s)^2S \rightarrow Li(1s)(2p)^2\ ^4P$. We reconsidered this problem in a more precise manner using the distorted wave theory. Our distorted wave calculation includes distortion of the initial and final states of the incoming and outgoing electrons separately by using different distorting potentials. We have used the static potential, the polarization of the target lithium atom as well as the exchange of the incoming electron with the bound electrons in the target. The bound states of the target are represented in initial and final states by the Hartree Fock wave functions as used by Kulander and Dahler.[4] The details of the analysis, results and discussion will be presented at the conference. In Table I we briefly display our results using various versions of the distorted wave theory (E_i is the incident energy in Ry, I and F represent the inclusion of the initial and final state static potentials respectively, E and P signify the additional inclusions of the exchange and polarization potentials).

TABLE I

Electron Excitation of $Li(1s)^2(2s)^1\ ^1S$ to $Li(1s)^1(2p)^2\ ^4P$

in Units of $10^{-4}\ \pi a_0^2$

E_i	IF	IFE	IFEP
4.6	0.0389	0.191	0.266
4.7	0.202	0.207	0.309
4.8	0.331	0.339	0.322
5.0	0.335	0.342	0.313
5.3	0.250	0.255	0.266
5.5	0.206	0.211	0.231
5.6	0.188	0.192	0.214
6.0	0.133	0.136	0.155
7.0	0.0609	0.0627	0.0690
8.0	0.0302	0.0312	0.0326

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*Visiting Professor at the University of Toledo for the Spring of 1986.

NOTES

WEDNESDAY, OCTOBER 22, 1986

MERCER III
9:00 AM–11:00 AM

II/III-WA1–4

JOINT SYMPOSIUM OF THE TOPICAL MEETING ON MULTIPLE EXCITATIONS OF ATOMS AND THE INTERNATIONAL LASER SCIENCE CONFERENCE

Thomas J. McIlrath, University of Maryland,
Meeting Cochair

MULTIPHOTON EXCITATION AND IONIZATION OF ATOMS

H. Jara, U. Johann, T. S. Luk, I. A. McIntyre,
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It has been conjectured that atomic motions fundamentally different from those presently observed may be driven under extreme conditions of irradiation. Indeed, it has been suggested[1-4] that ordered many-electron motions of outer-shell electrons could lead to enhanced rates of coupling from the radiation field to an atom. In order to study the conditions necessary for new atomic responses to occur and to determine the influence of possible damping mechanisms, the properties of ion charge state distributions,[5] electron energy spectra,[6] and harmonic radiation[7] produced by irradiation of atoms with ultraviolet radiation with different pulse lengths, wavelengths, and intensities have been investigated.

Recent experiments[8] have begun to explore the atomic response to intense 248 nm irradiation ($\sim 10^{16}$ W/cm^2) with subpicosecond pulses. Previous work, as cited in References (5) and (6), had been conducted at a few specific wavelengths in the range between 193 nm and 10.6 μm with pulse lengths of $\geq$ 5 ps duration. Since the dynamics of electron ejection from an atom can[9-12] involve a time scale significantly shorter than ~ 5 ps, it is expected[10] that the nonlinear coupling with intense radiation will be significantly modified if sufficiently short pulses in the subpicosecond domain are used.

For these measurements, two ultraviolet laser systems were available. One was a recently developed KrF* (248 nm) laser system[13] which produces pulses having a maximal energy of ~ 23 mJ with a pulse duration of ~ 0.5 ps. The second source used was an ArF* (193 nm) laser system[14] capable of producing a

~ 5 ps pulse with a maximal energy of ~ 40 mJ. The focusing system used in the studies of electron spectra[6,14] produced a maximum intensity of ~ 10^{16} W/cm^2 at 248 nm and of ~ 10^{15} W/cm^2 at 193 nm, respectively, in the experimental volume. The focusing lens was not corrected for spherical aberration and had, therefore, a focal diameter[10] of ~ 20 μm which limited the achievable laser intensity, but made the experiments relatively insensitive to the detailed spatial properties of the laser beam. The apparatus used for the measurement of the ion states[5] and the electron spectra[6] have been described previously. Both are time-of-flight type spectrometers; for the electron measurements a magnetic mirror collimated the electrons while the ions were extracted with a static electric field. The harmonic radiation was generated in a gas jet produced by a pulsed value and subsequently detected with a grazing incidence spectrometer and a multichannel detector.[16]

Overall, the results of the subpicosecond studies at 248 nm (1) provide the first observation in ion spectra of the removal of an inner-electron in a direct multiquantum collision-free interaction, namely, an electron whose principal quantum number (n) is less than that characterizing the outer-most shell of the neutral atom, (2) demonstrate that the pulse width is an important parameter in the coupling, (3) reveal the characteristics of the spectra of the energetic electrons up to ~ 250 eV formed by the interaction, and (4) exhibit the production of coherent radiation at 14.6 nm, the seventeenth harmonic of 248 nm and the shortest wavelength produced by that means.

Acknowledgements

The authors wish to acknowledge the technical assistance of R. Slagle, J. Wright, T. Pack, and R. Bernico. This work was supported by the U.S. ONR, the U.S. AFOSR, the SDIO(ISTO), the U.S. DOE, the LLNL, the NSF, the DARPA, and the LANL.

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Laser Spectroscopy of Core-Excited Levels of Neutral Rubidium

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We describe first experimental results on a new technique for obtaining level positions, autoionizing linewidths, and oscillator strengths of core-excited levels.

The technique is based on the large radiative rates, relative to their autoionizing rates, of levels that have recently been termed as quasi-metastable. Each of the column I metals and column II alkali-like ions have one or two such levels which radiate strongly to levels in the valence structure.

We excite a quasi-metastable level by hot electrons which are, in turn, produced by laser-generated x-rays, and view its radiation with a wide slit-low resolution monochromator. A tunable laser is scanned over the region where other autoionizing lines are expected to be present. When one is encountered, the quasi-metastable radiation is depleted, thereby determining its autoionizing linewidth and its position relative to the quasi-metastable level. By using a curve-fitting method in one-to-one correspondence with the curve-of-growth method, the transition oscillator strength may also be determined.

The method requires only a single (Quanta-Ray) Nd:YAG laser, used both to make the x-rays and to generate the tunable radiation. In rubidium, we observe radiation at 824 Å, which emanates from the even parity $4p^5 5s5p\ {}^4S_{3/2}$ quasi-metastable level. Thus far we observe depletion transitions to autoionizing lines at 148874, 150558, and 154018 cm^{-1}

relative to the Rb ground level. These levels have autoionizing linewidths of 1.3, 4.8, and 36.0 cm^{-1}, respectively. Two of these levels have been seen in XUV absorption by Mansfield and one with $J = 5/2$ has been seen in ejected electron spectroscopy by Pejcev, et al. These first results give linewidths and, therefore, autoionizing times, to an accuracy which is about twenty times greater than that given previously.

In an earlier experiment, using a pulsed hollow cathode, Pedrotti observed radiation at 207 Å in neutral lithium and used this depletion technique to obtain a width of 25.5 cm^{-1} for the lithium $1s(2s2p\ ^3P)^2P$ autoionizing line at 475163 cm^{-1}. The signal strengths in the present x-ray excitation technique are about twenty times greater than those obtained with the same element in a pulsed hollow cathode. Under typical conditions, we measure voltages which are equivalent to about 10 counts in a 5 ns interval. This allows scanning of a single line in under a minute.

In summary, this new technique allows the observation of autoionizing lines with both even and odd parity and with J values which make them inaccessible by optical absorption from ground. Linewidths are measurable to within laser accuracy and transition oscillator strengths within the core manifold may be determined. The observation of Rydberg transitions and interferences should also be possible.

The work described here is supported by the Air Force Office of Scientific Research and by the Army Research Office. K. D. Pedrotti is now at Rockwell International (1049 Camino Dos Rios, P.O. Box 1085, Thousand Oaks, CA 91360) and S. C. Wallace is at the University of Toronto (Department of Chemistry, Toronto, Ontario M5S 1A1, Canada).

AN OVERVIEW OF ABOVE THRESHOLD IONIZATION: THERE IS ONLY ONE PHYSICS

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The phenomena of Above Threshold Ionization (ATI) has been under intense investigation for nearly 10 years now. During this time not only have many diverse experimental and theoretical investigations been reported, but several major conferences have been held as well with the professed goal of achieving some consensus in the interpretation of the basic phenomena. Still, the ATI research community remains largely fragmented and confused about the basic physics and is in danger of dividing into separate "camps". In this paper I argue that the fundamental ideas necessary to understand the experimental results in ATI have already been put forward by many authors, often in other fields. For example, theorists interested in non-linear Compton scattering worked out the form of the interaction of high intensity light with electrons, and plasma physicists wrote down the consequences of this interaction for real-life measurements of kinetic energies of electrons produced in focused, high-intensity beams. Many investigators concerned with the non-linear interaction of high-intensity light on bound electron systems had to understand the concept of optical field ionization and have subsequently applied this knowledge to the measurement of spectra obtained in multiphoton ionization of molecules with ir light. I will show that if the relevant conclusions of these and other works are combined with the body of experimental and theoretical work from the ATI community, a rather simple and appealing description of ATI emerges, one which emphasizes the universality of its nature.

Multiple Excitation and Ionization of Atoms by Strong Lasers: Is There Any New Physics?

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Although, under certain conditions a transition probability per unit time averaged over the laser pulse may be useful in interpreting general features of experimental results, in general events during the evolution of the pulse must be followed in detail because total ionization of various ionic species is apt to occur.[1] As a result, at different stages of the pulse, processes of different order may appear. The end result is ionization. But a number of other processes such as wave mixing and lasing can coexist or even compete with ionization under the appropriate conditions. Employing calculated generalized cross sections, sequential ionization is calculated and shown to be the dominant mechanism under the conditions of recent experiments.[2,3] But even in sequential processes, multielectron excitations may play a role in determining the cross section and the state of the ion. Examples of calculations in multielectron atoms illustrate the role of more then one electron excitation. Doubly excited states, for example, can in

principle be employed in a sequence of transitions leading to a high lying doubly excited state. The probability of such transitions is estimated and the importance of various competing processes leading to sequential electron ejection is also discussed. In all cases, the structure of a particular atom and the frequency of the laser are found to play an essential role in the details of the process. Specific results pertaining to He, C, Sr and Xe are discussed as examples of the behavior of multielectron atoms in strong laser fields.

Supported by Grant No. PHY-8306263 from the National Science Foundation.

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NOTES

KEY TO AUTHORS, PRESIDERS, AND PAPERS

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Wallace, S. C. — II/III-WA2
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Zhu, Shitong — II-TuC6
Zhu, Y. — II-TuC10

TOPICAL MEETING ON

MULTIPLE EXCITATIONS OF ATOMS

POSTDEADLINE PAPERS

OCTOBER 20-22, 198
SEATTLE, WASHINGTON

1986 MULTIPLE EXCITATIONS OF ATOMS
MONDAY, OCTOBER 20, 1986
SEATTLE CENTER, MERCER III
II-MC POSTDEADLINE PAPERS

3:30PM II-MC1 Electron Spectrum in Intense-Field Photoionization, H. R. Reiss, University of Arizona and The American University.

A nonperturbative theory is applied to the intense field multiphoton ionization of xenon. Agreement with experiment is very good for high intensities and high orders.

3:45PM II-MC2 Multiple Charges Creation (Up to Xe^{6+}) from Xe Atoms by an Intense CO_2 Laser, S. L. Chin, W. Xiong, Université Laval, Canada, and P. Lavigne, INRS-Energie, Canada.

We observed ionization of Xe atoms creating ionic state of up to Xe^{6+} by a 1 ns CO_2 laser pulse at 10.6μ The results implied step-wise ionization.

4:00PM II-MC3 Subpicosecond Ultraviolet Multiphoton Electron Spectroscopy of Rare Gases, T. S. Luk, H. Jara, U. Johann, I A. McIntyre, A. McPherson, A. P. Schwarzenbach, K. Boyer, and C. K. Rhodes, University of Illinois at Chicago.

Photoelectron measurements of rare gases using subpicosecond 248 nm radiation focussed to the maximum intensity of $\sim 10^{16}$ W/cm^2 are described. Suppression of above threshold electron lines is observed and the interpretation is found to be consistent with the ponderomotive potential picture. In addition, line broadening and shift effects of the electron lines are examined.

4:15PM II-MC4 Multiphoton Double Ionization of Ba, Y. Zhu, U. Eichmann, and T. F. Gallagher, University of Virginia.

Using multiple laser excitations we have shown that Ba^{++} is formed via the 6s and 5d states of Ba^+.

4:30PM II-MC5 Double-Rydberg Spectroscopy of Barium Atom, P. Camus, P. Pillet, C.N.R.S. II, France and J. Boulmer, Universite de Paris-Sud, France.

Stabilization properties in 9dnd double-Rydberg series of barium are discussed. Selective microwave ionization techniques coupled with a double time-of-flight mass spectrometer are used to analyze highly excited Ba^+ Rydberg states.

Electron Spectrum in Intense-Field Photoionization

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A nonperturbative theory of multiphoton ionization was given in 1980[1] by the present author, which predicted the subsequently observed phenomena of above-threshold ionization (ATI), suppression of lower-order peaks in the spectrum of ionized electrons, and major qualitative differences in the spectra of electrons ionized by circularly polarized light as compared to linearly polarized light. The theory uses a short-range binding potential with a single bound state, much like the later independent work of Muller, Tip, and van der Wiel.[2] The 1980 theory is here applied to the case of photoionization of xenon by circularly polarized light of 1064 nm, for which experiments have recently been carried out.[3-5]

Comparison of theory with the experiments of Yergeau et al. is only moderately successful because of the low intensity involved. Both theory and experiment show suppression of the peak of minimal order at $n = 11$, they differ markedly on the peak at $n = 12$, and then show great similarity for the remainder of the spectrum. At the higher intensity used by Bucksbaum et al., the correspondence of theory and experiment

is much more striking. There is agreement that the peaks at n = 11, 12, and 13 are missing, the highest peak is at n = 17, and there is an excellent qualitative accord on the shape of the envelope enclosing the peaks.

Theoretical results are also presented from the theory at even higher intensities than those explored experimentally. At 1.1×10^{14} W/cm^2 with circularly polarized 1064 nm light, peaks from n = 11 to n = 24 are suppressed, and the bell-shaped envelope of peaks reaches its maximum at n = 34. It is also concluded that the ionization channel leading to a final $^2P_{1/2}$ state, which is of little importance at lower intensities as compared to the channel leading to the $^2P_{3/2}$ state, achieves more significance at high intensity.

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MULTIPLE CHARGES CREATION (UP TO Xe^{6+}) FROM Xe atoms BY AN INTENSE CO_2 LASER*

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Introduction

Experiments in recent years have shown that using an intense laser beam of intensities of more than 10^{13} W/cm^2 (up to 10^{16} W/cm^2) multiply charged ions of rare gas atoms were created with relative ease. The wavelengths of the lasers ranged from the infrared[1,2] through the near i.r.[3] and visible[3] to the u.v.[4]. Most of the data indicated that a step-wise multiphoton process was responsible for the multiple charge creation.

In the case of using a CO_2 laser[1,2], up to Xe^{3+} was created through a step-wise process although it was not yet clear whether each step involved a tunnelling or a multiphoton mechanism. The present work seeks to observe still higher charge states from ionizing Xe atoms using a CO_2 laser. The question we ask is if very much higher CO_2 laser intensities are needed in order to observed charge states higher than 3+.

The experimental method is the same as that used in our previous work for the ionization of rare gas atoms[1,2]. In order to increase the laser intensity, we used the highest gain 10.6μ laser line of 1 ns duration. The improved vacuum system was pumped down to a background pressure of 1 x 10^{-8} torr using a turbomolecular pump. Compared to previous work[1,2], impurity ions arriving in the time interval where Xe^{4+}, Xe^{5+} and Xe^{6+} would occur on the time of flight mass spectrum were significantly reduced so that we can now distinguish the various Xe ions from the impurities. A 25 cm focal length lens was used to further increase the laser intensity.

Results and discussion

Ions of up to Xe^{6+} were observed with relative ease from the time of flight signals. Unfortunately, the signals for Xe^{4+}, Xe^{5+} and Xe^{6+} were always too low to allow for a meaningful ions versus intensity plot. That was due to a temporary saturation of our high gain electron multiplier caused by the lighter impurity ions (mainly H^+ and H_2^+) when very high intensity was used. In spite of that, we were able to estimate the lowest intensities at which these highly charged ions start to appear. Fig. 1 shows a plot of the appearance intensity as a function of the ionic charged states. The appearance intensity was the lowest intensity at which a detectable signal was observed using our present set up at a Xe pressure of about 10^{-6} torr. Both Xe^{5+} and Xe^{6+} occurred in the same range of intensity except that statistically, Xe^{5+} appeared more often than Xe^{6+}. In fact, we observed that, on the average, for every four occurrences of Xe^{5+}, there were three occurrences of Xe^{6+}. This means that the appearance intensity of Xe^{6+} should be slightly higher than that of Xe^{5+} and this is shown as a circle on Fig. 1 at an arbitrarily slightly higher value.

Although Fig. 1 does not give an absolute measure of the ionization probability, it does show an important trend. Contrary to what one might imagine intuitively, the intensity at which higher charge states were created did not increase sharply as the ionic charge state increased. Instead, the appearance intensities flattened off rapidly from "4+" to "6+". This means that the probability of creating Xe^{6+} was rather close to that of creating Xe^{5+} and they are both not far away from that of creating Xe^{4+}. This seems to favor a step-

wise ionisation process in the creation of Xe^{4+}, Xe^{5+} and Xe^{6+}, on account of the following argument.

First of all, from Table I, we see that the ionization potentials from Xe^{4+} to Xe^{5+} and from Xe^{5+} to Xe^{6+} are rather close to each other (53 and 58 eV respectively). The ionization potential from Xe^{3+} to Xe^{4+} is not far away either (42 eV).

Secondly, assuming a step-wise process, because of the small photon energy ($h\nu = 0.117$ eV), each step of ionization involves, energetically speaking, the absorption of the radiation energy that correspond to more than a hundred photons at a time (see Table I). (The total minimum number of photons needed for the creation of Xe^{6+} in a step-wise process is thus 1862, a record number). Even if there is a difference of only a few eV (for example, 5eV between the ionization potentials for the step-wise creation of Xe^{5+} and Xe^{6+}), this corresponds to a difference of 43 photons (see Table I), meaning that the order of nonlinearity has increased by 43 if multiphoton ionization process were still involved.

The similarity of the ionization probabilities is hard to imagine at first glance. Yet, precisely because of such a high order non-linearity, the "ON-OFF" hypothesis[2] which we have proposed to explain the experimental data for the ionization of all the rare gas atoms using two arbitrary CO_2 laser lines (10.55μ and 9.55μ) could be applied here. Essentially, the "ON-OFF" hypothesis assumes that the probability of ionization of an atom through a highly non-linear process, be it multiphoton or tunnelling, becomes unity as soon as the intensity of the laser reaches a threshold value below which the probability is exactly zero. Thus, because of the highly non-linear processes involving non-linear orders of 453 and 496 respectively for the step-wise creation of Xe^{5+} and Xe^{6+}, a difference of 43 is not important any more and the ionization "threshold" should not be very different. Similarly, the difference in ionization potentials for the step-wise creation of Xe^{4+} and Xe^{5+} is 11eV, corresponding to about 94 photons. The difference in the appearance intensities is thus larger than that between Xe^{5+} and Xe^{6+}. If any of the processes for the creation of Xe^{4+}, Xe^{5+} and Xe^{6+} were due to a coherent direct process, the appearance intensity would have jumped up significantly which is not what we observed.

In conclusion, we have observed that up to Xe^{6+} have been created with relative ease using a 10.6μ 1 ns CO_2 laser pulse while the appearance intensities for the creation of Xe^{4+}, Xe^{5+} and Xe^{6+} are rather close to each other. A step-wise process coupled to the "ON-OFF" hypothesis could explain these qualitatively.

Acknowledgement

This work was supported in part by Le Fonds FCAR of Quebec and in part by the Natural Sciences and Engineering Council of Canada. The expert service of the Optical Component Laboratory in the Physics Division of the National Research Council of Canada is appreciated. We thank also the technical service of Marco and Pierre-Paul. One of us (W. Xiong) appreciates a graduate scholarship from the Ministry of Education of the People's Republic of China.

TABLE I

Ionization potentials (I.P.)
for the creation of multiphy charged Xe ions
(from Handbook of Physics and Chemistry, vol. -----)

Xe	1+	2+	3+	4+	5+	6+	
I.P.(eV)	12.127	21.2	31.3	42	53	58	
Corresponding photon number ($1h\nu$ = 0.117eV)	104	182	268	359	453	496	total 1,862

FIGURE CAPTIONS

Fig. 1 A plot of the appearance intensity versus ionic charge state of Xe created by an intense CO_2 laser pulse.

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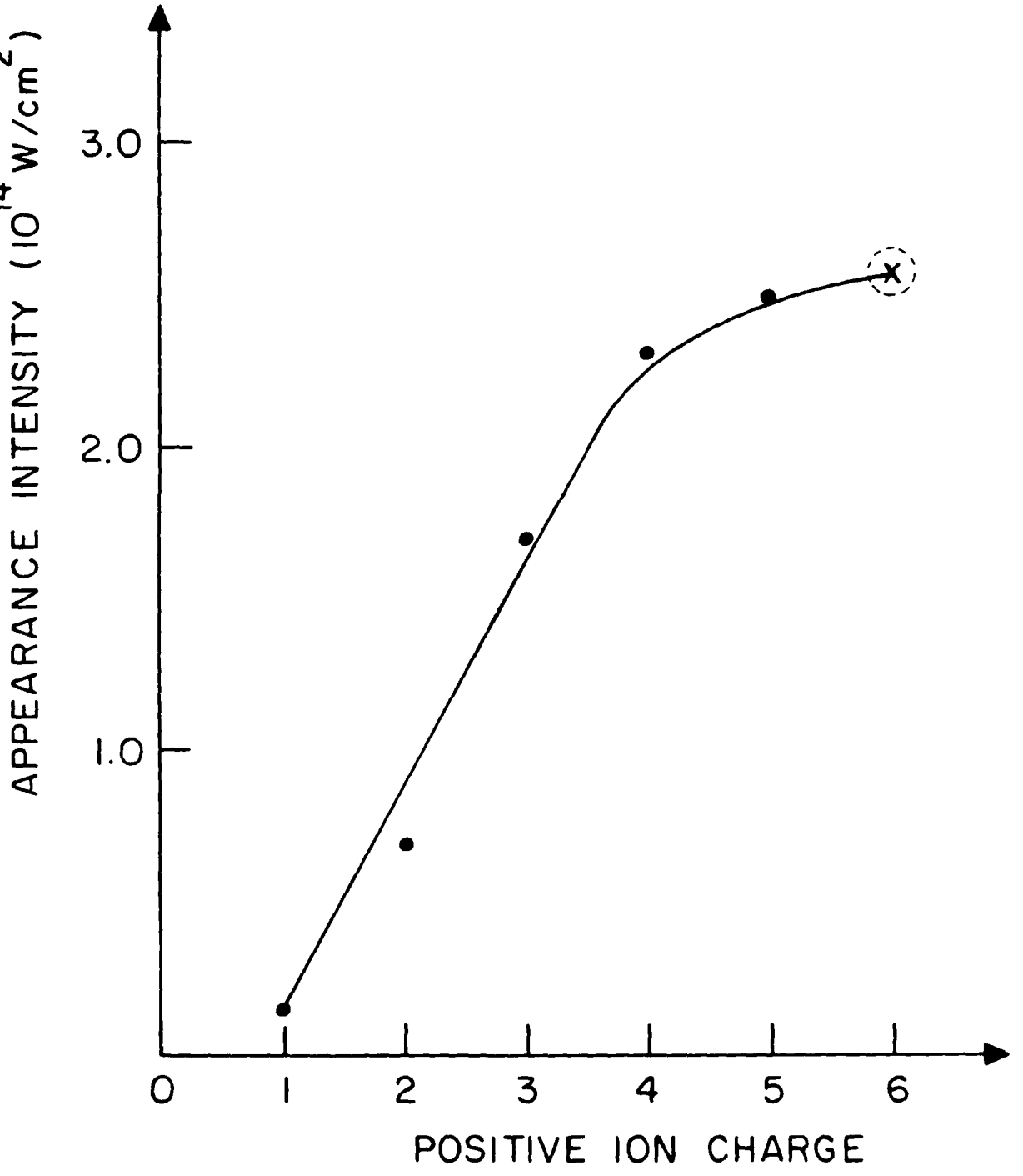
APPEARANCE INTENSITY (10^{14} W/cm^2)
3.0
2.0
1.0
0
1
2
3
4
5
6
POSITIVE ION CHARGE

MC3-1

Subpicosecond Ultraviolet Multiphoton Electron Spectroscopy of Rare Gases*

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Recent advances in high power laser development have enabled field strengths comparable to the atomic binding field to be generated. This experimental achievement has stimulated considerable interest in the study of the nature of multiphoton processes. Previously reported results of photoelectron spectra using 10 ps, 193 nm radiation[1] have shown that the photoionization mechanism for the low charge states is largely a stepwise process and the threshold intensities required to produce the charge states are in reasonable agreement with the Keldysh model. In addition, the effect of the ponderomotive potential in suppressing the lowest order ionization channel was observed.

In this experiment, a laser system[2] of much shorter pulse duration, 0.5 ps at 248 nm, has been used to study multiphoton coupling of the rare gases. Figure 1 shows the electron spectrum of He at 2×10^{14}, 4×10^{14}, and 2×10^{15} W/cm^2. The lowest order energy electron line, which should appear at 0.4 eV, is not shown. At ~ 1×10^{14} W/cm^2, however, there is a distinct electron line of ~ 5 eV which represents the second order line above the ionization threshold. As the intensity increases further, the 5 eV line is totally suppressed. This result is the first observation of the suppression effect for the above threshold ionization (ATI) lines other than the lowest order.

There are three other features worth noting; (1) the electron lines are shifted to lower energy, (2) the line width of the electron lines are broadened significantly, and (3) electron energy as high as 250 eV in Xe and Ne have been observed. The interpretations of these features will be presented.

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Fig. (1): Electron energy spectra using 0.5 ps 248 nm laser focussed with 21 cm focal length, (a) 2 x 10^{14} W/cm^2 at 1 x 10^{-5} Torr, (b) 4 x 10^{14} W/cm^2 at 2 x 10^{-7} Torr, and (c) 2 x 10^{15} W/cm^2 at 2 x 10^{-7} Torr.

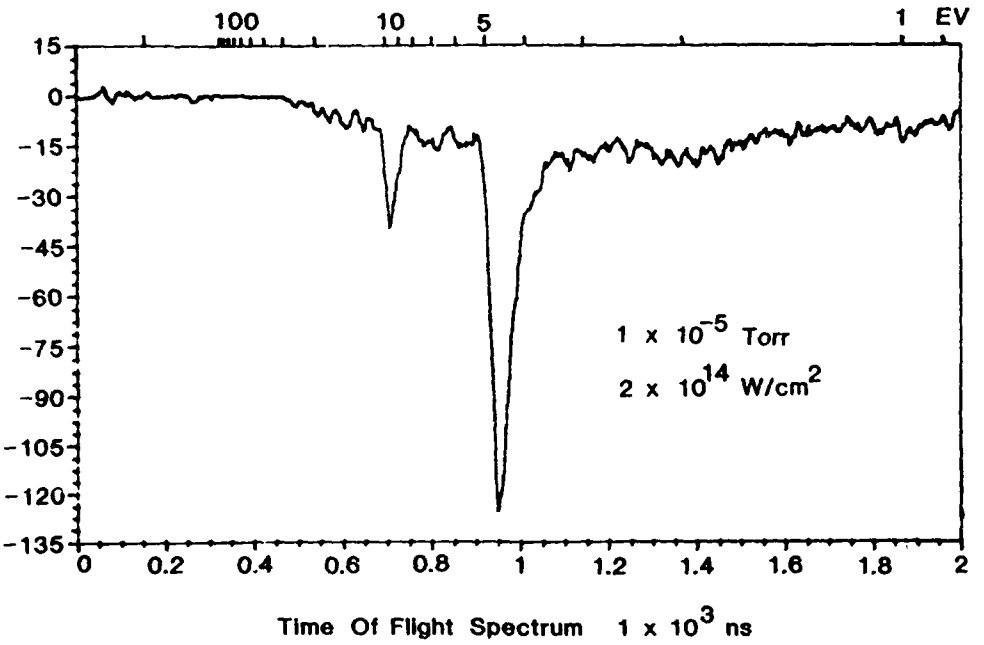

Figure 1a.

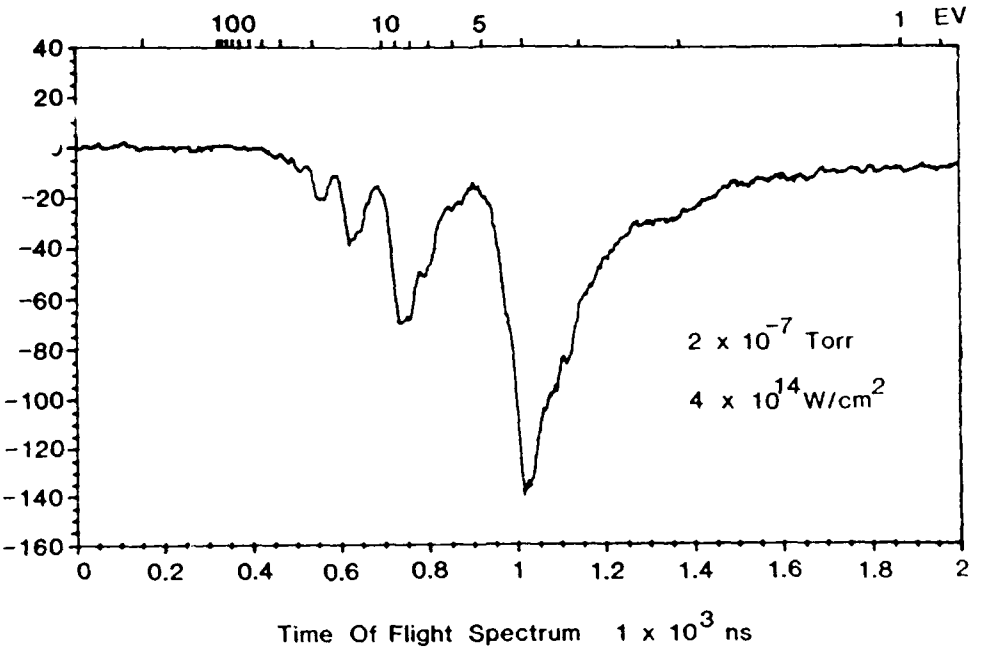

Figure 1b.

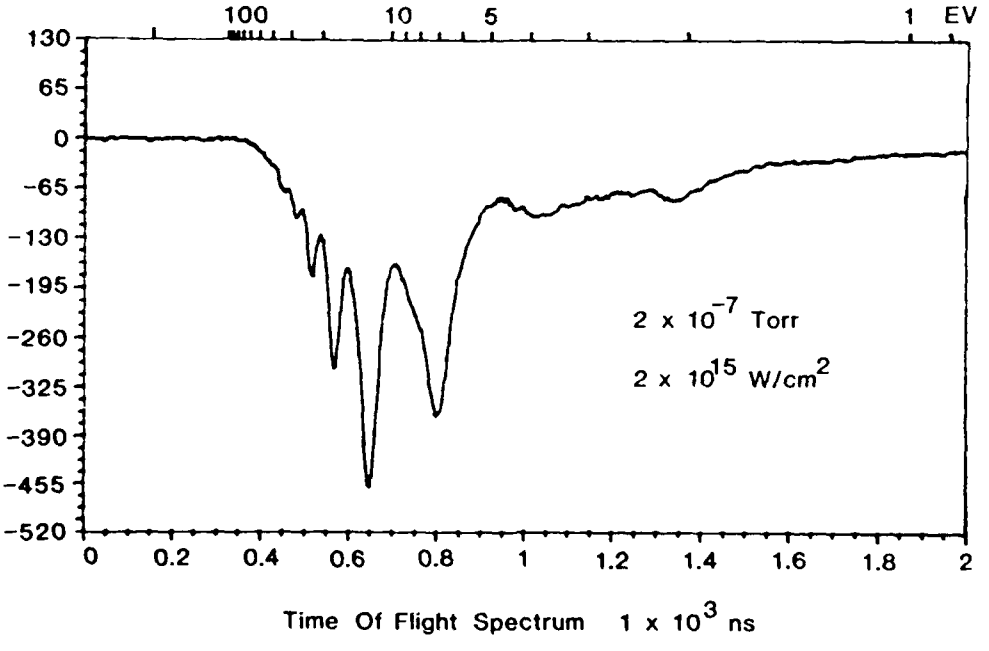

Figure 1c.

II-MC4-1

Multiphoton Double Ionization of Ba

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In an effort to gain more insight into the process of multiphoton multiple ionization,several experiments have recently been carried out to determine the wavelength dependence of the double ionization of Sr and Ba.[1-3] These studies were not as conclusive as one might hope, in that the observed resonances in the production of Sr^{++} and Ba^{++} did not quite match the energy levels of Sr^{+} and Ba^{+}. Here we report experiments in which we have compared the spectra for the production of Ba^{++} by an intense laser with and without the population of specific states of Ba^{+} by modest secondary lasers. The results of these experiments show clearly that Ba^{++} is produced by first producing real intermediate states of Ba^{+} which are multiphoton ionized through intermediate resonances in Ba^{+}.

In these experiments a thermal beam of Ba is crossed by several laser beams; an intense beam of 50 mJ pulse energy and 5ns pulse duration which is focussed to a diameter of 0.1 mm with a 100 mm focal length lens, and two modest beams of 1 mJ pulse energy. After the laser pulses a voltage is applied between plates above and below the focal volume to eject the Ba^{+} and Ba^{++} ions, which are readily identified on the basis of their flight times to the detector.

First the Ba^{++} spectra are recorded as a function of the wavelength of the intense laser without the secondary lasers. Then the spectra are recorded with the secondary lasers which are timed to precede the intense laser by 100 ns. The secondary lasers both subtantially deplete the ground state of Ba and produce a real population in a specific state of Ba^{+}. Spectral features which are diminished by the addition of the secondary lasers do not originate in either the ground state of Ba or the selected state of Ba^{+}. On the other hand features which increase upon the addition of the

secondary lasers must originate in the selected state of Ba^+.

These observations show clearly that the double ionization of Ba from 550 to 670 nm proceeds through real intermediate states of Ba^+, specifically the $6s_{1/2}$, $5d_{3/2}$, and $5d_{5/2}$ states. The locations of the observed resonances reflect the intermediate Ba^+ resonances in the multiphoton ionization of the low lying states of Ba^+.

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II-MC5-1

DOUBLE RYDBERG SPECTROSCOPY OF BARIUM ATOM

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One of the interesting problem of atomic physics is to understand better the dynamic of highly excited two-electron systems ($n\ell n'\ell'$). Recently, the exploration of highly excited two-electron states so called "double-Rydberg" states have been performed using multiphoton laser excitation in barium. From a theoretical point of view two classes of "double-Rydberg" states can be considered. At the so-called Wannier ridge ($n \backsim n'$), the correlation between the electrons becomes predominant. On the other hand, on both sides of the Wannier ridge ($n' \gg n$), the outermost electron is moving in the field of the ionic core, partly screened by the inner electron. Properties of such "double-Rydberg states called "valley states" or planetary atoms could be described in terms of quantum defect theory.

SPECTROSCOPIC STUDY OF 9dnd DOUBLE-RYDBERG STATES

Properties of the neutral doubly excited states, mainly 9dn'd (n'=17 to 29) double-Rydberg states have been studied using a two-step two-photon laser excitation, via 6snd $^1D_2$ simple Rydberg state. These states autoionise in $Ba^+(m\ell)$ ($m<9$) and then the produced ions are photoionised in Ba^{2+} ions. A time of flight mass spectrometer is used to discriminate and detect the Ba^{2+} ions. The two autoionizing Rydberg series $9d_{3/2}n'd$ and $9d_{5/2}n'd$ have been observed and interpreted in the analytic model of a two-channels quantum defect theory. Most of the lines are explained and understood in this simple model. Nevertheless, some new features which are due, for instance, to the quasi-coincidence and the interference between two members of the two series $9d_{3/2}29d$ and $9d_{5/3}27d$ leads to a narrowing of the autoionizing width which corresponds to a "stabilization" of the resonance. A theory with six channel quantum defect analysis take into account mixing between the series and allow us to fit the experimental data.

TOWARD HIGHLY EXCITED DOUBLE-RYDBERG STATES

Higher excited double-Rydberg states of Ba atom autoionize in highly excited Rydberg of Ba^+ ion. These products of autoionization shall be detected and analysed for studying double-Rydberg. A new method of detection, efficient and selective, has been performed by using microwave ionization coupled with a double time of flight mass spectrometer. This new technique allow is to observe the Rydberg series ns and nd ($20 < n < 65$) of Ba^+ ion.

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